Chemical Behavior of Iodine-131 during SRE Fuel Element Damage in July 1959

Response to Plaintiff's Expert Witness Arjun Makhijani

by

Jerry D. Christian, Ph.D.

Prepared for *in re Boeing* Litigation

May 26, 2005

Background of Jerry D. Christian

Education:

B. S. Chemistry, University of Oregon, 1959.

Ph. D. Physical Chemistry, University of Washington, 1965 – Specialty in Chemical Thermodynamics and Vaporization Processes of Halogen Salts. (Iodine is a halogen.)

Postdoctoral: National Research Council Senior Research Associate, NASA Ames Research Center, Moffett Field, CA, 1972-1974.

Career Summary:

Scientific Fellow, Retired from Idaho National Engineering and Environmental Laboratory (INEEL), September 2001. Scientific Fellow is highest achievable technical ladder position at INEEL; charter member, appointed in January 1987.

Consultant and President of Electrode Specialties Company since retirement.

Affiliate Professor of Chemistry, University of Idaho; I teach a course in nuclear fuel reprocessing.

Referee for Nuclear Technology and Talanta journals; I review submitted technical manuscripts for the editors for scientific and technical validity and accuracy.[•]

I have thirty nine years experience in nuclear waste and fuel processing research and development. Included in my achievements is development of the highly successful classified Fluorinel Dissolution Process for advanced naval fuels that was implemented in a new $250 million facility at the ICPP in the mid-1980s. Career interests and accomplishments have been in the areas of nuclear fuel dissolution, aqueous fluoride chemistry, metal halide vaporization processes, high temperature ruthenium chemistry, solvent extraction separations chemistry, radioactive incinerator off-gas treatment, radioactive airborne waste management technologies, beneficial reuse of radioactive contaminated metals, oxidation of metals, thermodynamic modeling, activity coefficient calculations and models, technetium-molybdenum separations processes for medical isotope production, and characterization and simulation of radioactive sodium-bearing waste.

[•] Iodine technology manuscripts that I have refereed for Nuclear Technology include:

- "Kinetics of Gaseous Iodine Uptake onto Stainless Steel during Iodine Assisted Corrosion," 2000.
- "The Relationship between Molybdenum Oxidation and Iodine Volatility in Nuclear Fuel," 2003.
- "On the Radiolytic Decomposition of Colloidal Silver Iodide in Aqueous Suspension," 2004.

The first two are directly related and applicable to aspects of the SRE evaluations.

My work on ruthenium chemistry changed prevailing concepts in the scientific community about its behavior and chemistry during evaporation and calcination of high-level radioactive waste solutions and has influenced research on this topic around the world. My technical work has resulted in four patents, five books and book chapters, and numerous peer-reviewed journal articles, technical documents and reports, and technical presentations and lectures.

I received a Special Award from the Chairman of Westinghouse for development of the Fluorinel Dissolution Process for advanced naval fuels, was one of ten annual international Westinghouse Signature Award winners in 1993 – for a chloride volatility processing concept for spent nuclear fuels – and received the 1994 American Nuclear Society Special Award for Innovations in Long-Term Storage of Spent Nuclear Fuels. The Idaho Academy of Science awarded me their inaugural Distinguished Scientist/Engineer Award in 2000 "for an outstanding career of pioneering scientific research and leadership of national and international impact".

A full C. V. is attached.

Prior Testimony

I was retained by the defense to prepare an expert report in re: Hanford Nuclear Reservation Litigation, Master File No. CY-91-3015-WFN, United States District Court Eastern District Court, and was deposed in that case on October 26, 2004. I did not testify at trial.

Compensation

My compensation in this case is $300 per hour.

TABLE OF CONTENTS

FIGURES

TABLES

WHAT I DID

I was asked by the defendant in the Boeing litigation to evaluate issues concerning iodine behavior and iodine chemistry as they relate to the July 1959 SRE failed fuel incident.

I reviewed a number of documents related to the reporting and assessment of the fuel cladding failure incident and was also provided with opinion papers written by expert witnesses for the plaintiffs. I was asked to evaluate this material and the specified claims and statements made by the plaintiffs' expert Dr. Arjun Makhijani. I was also provided with a number of relevant SRE documents and reference reports of the witness Makhijani to aid in my reviews. During the course of my work, I supplemented this material through my own research and conversations with other experts in the field, as well as individuals with knowledge regarding sodium-cooled reactor operations and related issues. I included reviews of pertinent scientific literature.

I used my knowledge and expertise in iodine chemistry, in physical chemistry, and in nuclear fuel processing to evaluate the SRE incident. My background involves both scientific research and practical process development that includes implementation of a nuclear fuel dissolution process at the Idaho Chemical Processing Plant. These enabled me to assess the technical information and the behavior of iodine in the SRE.

I. INTRODUCTION AND OVERVIEW

A. Objectives and Organization of the Report

The primary objective of this study is to assess the events leading to and consequences of the July 1959 fuel cladding failures in the Sodium Reactor Experiment (SRE) that was fueled with uranium metal fuel. Of particular interest and emphasis is the fate of the iodine-131 (I-131) that was in the failed fuel elements. This is necessary in part because the plaintiffs' expert witness Dr. Arjun Makhijani asserts that some 34 percent (probable value) of it was released from the stack. The primary purpose of the current study is to provide an objective, independent evaluation of the fate of the iodine and the source term for release from the stack. The evaluation considers operating and sample data collected from the incident; pertinent chemical effects, starting with the fuel and continuing to the path to the stack; relevant scientific literature that includes chemistry of iodine in the fuel, experiments measuring retention of iodine released in bubbles into sodium, observations of iodine behavior during similar metal-fueled, sodium-cooled reactor fuel failures, and experimental characterization of the amount and chemical form of iodine released from heated and melted metal irradiated fuels.

When there are differences in conclusions with those of the plaintiffs' expert witness, further discussion is needed to explain those differences. In every instance, scientific considerations and detailed assessments of plant design and operating data do provide explanations. Thus, following my evaluations, a section is added that assesses Dr. Makhijani's assertions in light of scientific principles, experimental data, and plant design and operating data. Because it is difficult to always separate the independent review and the need to answer specific associated questions raised by Dr. Makhijani, the review does include an occasional reference to and clarification of Makhijani's claims. But, the independent review is focused on facts of the incident. Evaluations of Dr. Makhijani's specific claims are reserved for separate analysis, where possible.

The evaluation of the SRE incident is, necessarily, highly technical in nature and often involves consideration of details that can sometimes be somewhat laborious to follow. It is essential that all assessments and arguments include and address all known facts and scientific derivations that are pertinent, rather than to make qualitative observations, suggestions, and claims without detailed quantitative analysis. However, in an attempt to make the discussions readily followed and understood, the very most detailed and technical derivations and analyses are moved to appendices with the overall conclusions summarized in the main text. In this way, the reader can and should verify the statements made in the text. Still, much technical information is provided in the text because it is the only way for the reader to immediately follow and understand the line of reasoning.

The order of assessments is to, first, review relevant literature data regarding release of fission products from metal fuels during normal operations of reactors and from failures of metal fuels in reactor fuel failure occurrences and literature reports of experimental investigations characterizing fission gas releases from metal fuels under various conditions. This provides a background basis for assessing the situation and for understanding data collected during the SRE incident. Then, the operating and other reported data from the SRE July 1959 incident are assessed in a quantitative fashion. Finally, literature data relevant to the behavior of iodine in the sodium environment at various stages of the reactor and off-gas system are discussed in light of what would have happened to any molecular iodine if it were released from the fuels as asserted by Dr. Makhijani.

B. Overview

The Sodium Reactor Experiment (SRE) was a sodium-cooled, graphite-moderated, thermal power reactor fueled with slightly enriched uranium metal with uranium enriched to 2.78% uranium-235.[1, p. 2], [2], [3], [4, p. 74] It was designed and constructed by Atomics International, a division of North American Aviation, Inc., as part of a larger program with the Atomic Energy Commission to develop a civilian, sodium-cooled, thermal power reactor.[5] It was constructed on a 44 acre site in the Santa Susana Mountains – 30 miles west of Los Angeles and 10 miles from Atomics International headquarters in Canoga Park, California.

On approximately July 12, 1959 and thereafter tetralin that had leaked into the sodium coolant reacted to form a carbonaceous particulate material to an extent that restricted the flow of coolant around some fuel channels and resulted in overheating of some of the fuel elements in the core of 43 elements of the SRE reactor. The increased temperature with uranium fuel slugs in contact with the stainless steel cladding caused rapid diffusion of uranium into and alloying with the stainless steel. At locations where the temperature exceeded the melting point of the iron-uranium eutectic, 1337°F (725°C), this diffusion resulted in the formation of an alloy with some liquid phase present. This alloying ultimately resulted in failure of the cladding of some of the fuel elements, though the fuel did not melt. Over the course of the next several days, cladding of 13 of the 43 elements failed. Of the 13 failed element claddings, three showed no evidence of eutectic melting. However, thermocouples showed that they cycled several times through the α-β phase transformation temperature for uranium of 1225°F (663°C). A 5% anisotropic volume increase results during the change from the orthorhombic α phase to the tetragonal β phase and likely ruptured the cladding from swelling.

The maximum temperature achieved in the fuel elements was less than 1999°F (1093°C) and was likely no more than 1472 to 1652°F (800 to 900°C). The melting temperature of uranium, 2075°F (1135°C), was not reached.

An assessment of reactor data obtained during the incident shows that most of the failed fuels were damaged and released the observed small amounts of radionuclides in the sodium and in the cover gas on or about July 23. While a small amount of failure may have occurred in the July 13-July 15 period, the preponderance of evidence indicates that the majority of failures likely occurred between July 21 and 24. The plaintiffs' expert witness' basis for the iodine-131 source term is that most of the failures occurred with iodine and noble gas releases by July 15 so that he can postulate a release mechanism in which the cover gas with the fission gases was quantitatively swept by the holdup tanks on July 15 and to the stack during venting operations. While this failure date for most of the damaged fuels is highly unlikely, the present analysis examines what the consequences of that postulated scenario would have been. The analysis shows that, in fact, no iodine-131 could have been released from the failed fuels to the stack via Dr. Makhijani's postulated sequential mechanisms. The multiple reasons are explained in quantitative fashion and include fuel chemistry that shows iodine was present, not as gaseous elemental iodine, I_2, but as non-volatile uranium triiodide salt, UI_3, fission gas behavior at the very low burnup of the SRE fuels that prevented their releases from the damaged fuels, the postulated release path that would have removed iodine, cover gas and stack gas sample data that show negligible iodine, and detailed analysis of the cover gas venting data showing little transfer of any iodine out of the core cover region.

As explained in the text below, the incident did not result in significant release of any fission products, including gases, from the failed-cladding fuels. Of the small quantities released from the fuel, most, including all of the released iodine, were trapped in the sodium. Only small fractions of xenon and krypton escaped from the fuel and through the sodium into the cover gas. Xenon and krypton are not soluble in or chemically reactive with sodium. About 1% or less of failed element fission product inventory of non-volatiles, including iodine as a salt, was found in the sodium. No iodine-131 was found in the cover gas. About 4.3% and 0.062% of the xenon-133 and krypton-85 gases, respectively, were found in the cover gas, assuming that the bulk of the fuels failed about July 13. This ratio of 70 between the percent releases of xenon and krypton is not plausible and argues against this release date. If the fuels failed later in the run, the corresponding xenon and krypton releases were 0.18% and 0.038%, more in line with expectations of similar fractions released. The differences between the percent releases of the two gases based on a July 23rd release from fuels can be resolved if one considers a likely error in measuring krypton-85 by a factor of 4.7, which would simultaneously make the total activity concentration in the sample calculated for August 12 exactly equal to the gross activity measured independently for the sample. This kind of correlation cannot be derived for the factor of 70 difference in percent releases of krypton and xenon based on an assumed July 13th release from fuels.

The fate of I-131 during the incident is evaluated from a number of aspects including 1) assessment of data obtained prior to, during, and following the incident: analysis of the sodium coolant; analysis of the cover gas; 2) experiments characterizing iodine behavior in sodium, 3) experiments on irradiated metal fuel elements which were taken to failure, 4) data and experience from other sodium-cooled reactors during fuel meltdown failure and during normal operations with cladding failures, 5) thermodynamic modeling of fission products in uranium metal fuels, 6) assessments of venting of the cover gas on July 12th and 15th, and 7) stack monitoring data. The conclusion from all these considerations is that fission product iodine formed uranium triiodide and/or cesium iodide in the metallic fuel and was not released from the fuel as a gas. Based on considerations of the chemistry of iodine in the fuel that would make it behave similarly to other non-volatile fission products, on I-131 measured in the sodium, and on the lack of I-131 in the cover gas, only between 0.3 and 1.3 percent (depending on the assumed date of release) of the iodine-131 was released from the failed fuel elements. Of that released, all was captured and retained in the sodium coolant. No iodine was released to the stack. Details of the analyses are provided in the report.

Plaintiff expert witness Dr. Arjun Makhijani makes a number of qualitative observations regarding cover gas purges, sample gas decays prior to analyses, and stack monitoring to develop conjectures that the gases xenon, krypton, and iodine were released from the fuels and to the stack without being measured. However, he did not perform detailed analysis of all this information to quantify his observations. When detailed quantitative assessments are made of all data, even with the uncertainties raised by Dr. Makhijani, clear, conclusive evidence is developed that, in fact, proves that, first, only about a percent or less of the xenon and krypton inventories in the failed fuels were released from the fuels and to the stack and, second, no iodine was released to the cover gas and, subsequently, to the stack.

At the time of the incident, the Atomics International researchers could not explain data showing unexpectedly very small release fractions of gaseous fission products. The gaseous fission product category considered by them included iodine, along with xenon and krypton. Given the absence of scientific knowledge about the chemistry of metal fuels, it was not recognized at the time that 1) iodine would exist as a non-volatile metal iodide in the metal fuel and not as free elemental iodine (I_2) and 2) gaseous fission products have very small migration rates from the fuel before it has melted and, especially, at the low burnup of the SRE fuels at the time of the incident they would not agglomerate into bubbles that could migrate out at all. The fuel did not melt.

No measurements were made of residual fission products in the failed fuel elements in order to achieve mass balances for them. Had they been made, the results would have shown that almost all of the iodine-131 (I-131) inventory remained in the fuel following the cladding failures. Even though extensive data (laboratory and sodium reactor operating data) and thermodynamic considerations exist today, as explained in the text below, to support these two points, plaintiff expert witness Makhijani clearly is not aware of them or chooses to ignore them and uses the limited, i.e., incomplete, measurement data following the cladding failures to devise an incredible release mechanism for the I-131. He did not consider or evaluate whether the iodine might have been contained in the fuel. Furthermore, his scenario for iodine escape focuses on getting it through the sodium into the cover gas. His claim that experiments showing that elemental iodine gas released in inert gas bubbles in sodium does not reach the gas space above the sodium are invalid and do not represent conditions in the SRE is shown to be incorrect and, in fact those experiments bracket the SRE conditions. Even more significantly, he does not address the subsequent barriers of iodine in the cover gas being in direct contact with the sodium pool and vapors for one and one-half or more days during which it would have reacted with sodium to form sodium iodide salt and, then having to pass through a sodium vapor trap on its way to the stack, which would further have resulted in complete reaction of sodium with and removal of iodine from the gas.

Analysis of the data shows that had I-131 been released to the cover gas as hypothesized by Makhijani, it would have been observed in the August 12 cover gas sample along with the observed xenon-133 (Xe-133), since the I-131 would have been present at a larger activity concentration than Xe-133 at the time of the measurement. I-131 was looked for and not found. Contrary to Dr. Makhijani's assertion that I-131 was not detected because any I-131, along with Xe-133, would have undergone a combination of decay and flushing out prior to taking the August 12 cover gas sample and measuring its gamma spectrum on September 14, the quantitative analysis of the venting data, decay fractions, and the gamma signal measurements show that it would, in fact, have been present and measured on September 14 if any had been released from the fuel to the cover gas.

Dr. Makhijani's assessments are incomplete and do not incorporate and explain all known facts that would be necessary in order for a hypothesis to be credible. He is very selective of available information that he chooses to present, ignoring numerous data and scientific literature that counter his arguments.

Indeed, even if the iodine had escaped to the cover gas along with xenon and krypton in the percentages of failed fuel inventories suggested by Makhijani in the July 12-13 time period and been transported to the stack during July 12 and 15 pressure ventings, which it did not, stack gas activity measurements indicate that much less than about 10 Ci of total activity was released to the stack, of which less than 12%, or 1 Ci, would have been I-131.

II. DESCRIPTION OF SRE REACTOR

The Sodium Reactor Experiment was a small, 20,000 kilowatt (20 megawatt)-thermal experimental thermal nuclear reactor that was fueled with uranium metal rods. The original plans provided that reactor heat be dissipated to the atmosphere in air-cooled heat exchangers. Later, a parallel-connected steam plant was provided to generate electrical power. Installed by the Southern California Edison Company, the steam plant could produce approximately 6,500 electrical kilowatts with the reactor operating at its design power level of 20,000 kW thermal. This is the typical (33%) efficiency of power reactors. For perspective of the size, a modern nuclear power plant produces over 1000 MW of electrical power, so the capacity of the SRE was 0.6% of a modern power plant's capacity.

In order to understand the processes that resulted in fission product releases during the fuel cladding failures and the measurements that were made of radioactivity in various locations following the occurrence, a brief description of the salient feature of the reactor is given here.

The uranium fuel was in the form of long, thin rods, each with twelve slugs of 0.75 inch diameter by 6 inch long slugs. The slugs were encased in 0.010 inch thick 316 L stainless steel tubes to form the rods. A liquid metal, sodium-potassium alloy (NaK), filled the 0.010 inch gap between the uranium slugs and the cladding to effect heat transfer from the uranium to the cladding. It may be noted that the NaK is similar to sodium (Na) in that it would react with any iodine that may be released from the uranium to form sodium iodide and potassium iodide, which are salts similar to table salt. (However, as described in detail in the subsequent sections, elemental iodine (I_2) was not released from the uranium.) Although there was an annulus (gap between two concentric cylinders) between the uranium slugs and the cladding, the slugs were loose in their jackets, some were bowed and sections of the uranium slugs would contact the stainless steel tube. Furthermore, by the time of Run 14 when the coolant blockage occurred, radiation-induced swelling had occurred to the extent that the fuel was scheduled to be changed out following Run 14 and resulted in more extensive contact of the uranium with the cladding.[6, p. II-B-8]

A fuel element was comprised of seven of these fuel rods in the form of a hexagon of six rods with the seventh in the center. Depictions of a fuel element are given if Figure 1. There were 43 elements in the reactor core. The total uranium loading was about 2.97 metric tons (2.97×10^6 g).

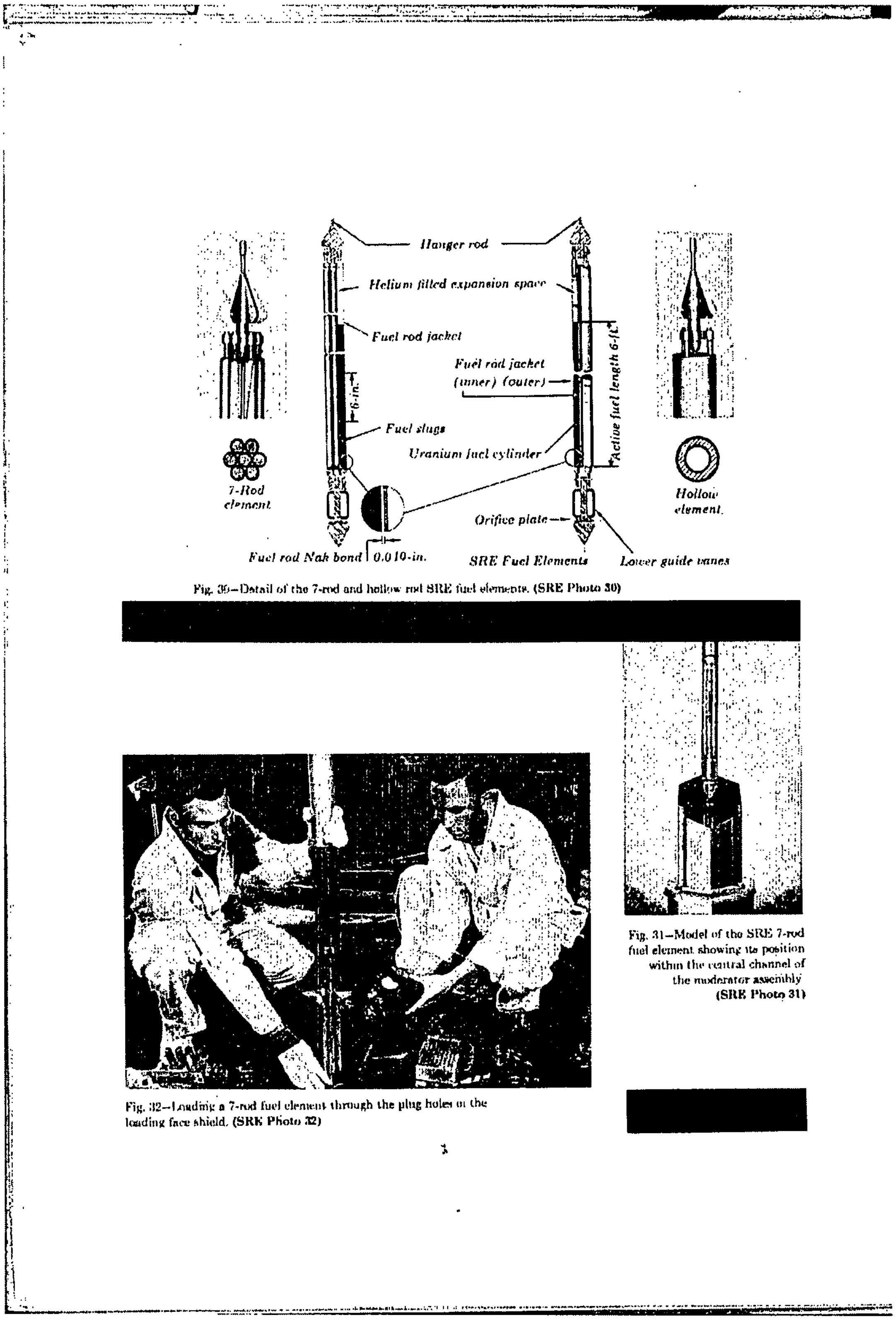


Fig. 30—Detail of the 7-rod and hollow rod SRE fuel elements. (SRE Photo 30)

[redacted]

Fig. 31—Model of the SRE 7-rod fuel element showing its position within the central channel of the moderator assembly (SRE Photo 31)

Fig. 32—Loading a 7-rod fuel element through the plug holes in the loading face shield. (SRE Photo 32)

[redacted]

Figure 1. SRE fuel element showing seven rods in the element. [5]

When Run 14 started, the burnup of the core was at 2409.7 MWd and at the shutdown, 2,425.8 MWd, corresponding to 811 and 817 MWd/t.[1, p. 7] Burnup, on a percent basis, is the amount of uranium that has fissioned, relative to the original total uranium present. It is sometimes expressed, also, in terms of the amount of energy produced per ton of initial uranium, i.e., MWd/T (megawatt days per short ton) or MWd/t (megawatt days per metric ton). The amount of energy produced per fission is 200 MeV, which corresponds to 0.948 MWd of heat produced per gram of uranium-235 fissioned.[7, pp. 54 and 88]. (Another equivalent expression of this is that 1.05% of uranium fissions per 10,000 MWd/t burnup.) Thus, at the 2,425.8 MWd for the core at the end of Run 14, the total uranium burned to the end of the run was 2,425.8/0.948 = 2,559 g, which is 0.086% of the 2.97 million grams of uranium core, that is, 0.086% burnup.

This is the average burnup for the core. Individual assemblies had slightly varying exposures. Actual burnups for individual assemblies have been compiled by Daniel & Associates[8]. Those for the thirteen failed elements at the end of Run 14 are compiled in Table I. Included is a column in which the assembly burnup has been increased by 10% to allow that an individual slug in a rod of the assembly would be exposed more than the overall average. On this basis, all but one have burnups of 0.1% or less, the single exception being R-43 (a mixed element) at 0.13%. The average for all thirteen failed elements is 0.084%. A working value of 0.1% is reasonable.

Table I. Burnups of the thirteen damaged fuel assemblies at the end of Run 14.

		Burnup		
Core Channel	Fuel Type	MWd/t	Percent	1.1×Percent
R-10	Std	645.2	0.066	0.075
R-12	Std	747.8	0.079	0.086
R-21	Std	853.0	0.090	0.099
R-23	Std	801.3	0.084	0.093
R-24	Std	303.5	0.032	0.035
R-25	Std	708.9	0.074	0.082
R-31	Std	745.1	0.078	0.086
R-35	Std	543.2	0.057	0.063
R-43	Mixed	1,144.4	0.120	0.132
R-55	Mixed	732.8	0.077	0.085
R-68	Std	870.3	0.091	0.101
R-69	Std	880.2	0.092	0.102
R-76	Std	518.3	0.054	0.060
AVERAGE		*730.3*	*0.077*	*0.084*

This will be seen as important in the following evaluations of fission product gas releases from the fuel where the percent burnup is seen to be a significant consideration.

The sodium pool above the reactor's fuel elements was covered by helium at a nominal pressure of between 0.5 and 3 psig (15.2 and 17.7 psia)[*],[2, p. 2, 9, pp. V-1 and V-4] an inert gas, to exclude air that would react with sodium. This helium was called the cover gas. It was normally operated at 1 psig.[9, p. I-A-2] During Run 14, it was operated at 2 psig. A schematic of the reactor showing these features is shown in Figure 2. The diameter of the sodium pool was 11 feet.[10, Fig. 2-1] The core cover gas volume varied, depending on the pressure above the sodium and the temperature of the sodium pool. Based on data tabulated by Hart, Table VII, for concentrations and total cover gas inventories of Xe-133 and Kr-85 gases sampled on August 12, 1959[1, p. 14], it was about 6,300 L on that date (corresponding to a depth of 28 inches), for purposes of calculating quantities of Xe-133 and Kr-85 based on different calculated source terms and modeled release dates. See Appendix B for additional description.

A shield with plugs was installed above the reactor core cover gas area that provided radiation shielding and access to elements in the core. The loading portion of the shield contained stepped casings for 81 small plugs. "O" rings provided a gas seal between the small plugs and casings. A low-temperature-melting alloy, cast into troughs at floor level, was used as the gas seal between the ring shield and the surrounding foundations and between the ring shield and the loading face shield. This plug assembly generally sealed the cover gas, although a very tiny amount of leakage could occur and this resulted in a very small amount of radioactivity, corresponding to a tiny fraction of the fuel fission gas inventories, measured in the room above the shield during the fuel cladding failures.[†] This room was called the reactor room or high bay. Schematics of the shield and of the reactor facility showing the high bay are shown in Figures 3 and 4, respectively. The volume of the high bay was 255,000 ft^3, based on the stated dimensions.[11, p. V-57] (Page B-2 of ref 11 uses the value 300,000 ft^3, probably as a rounded number.)

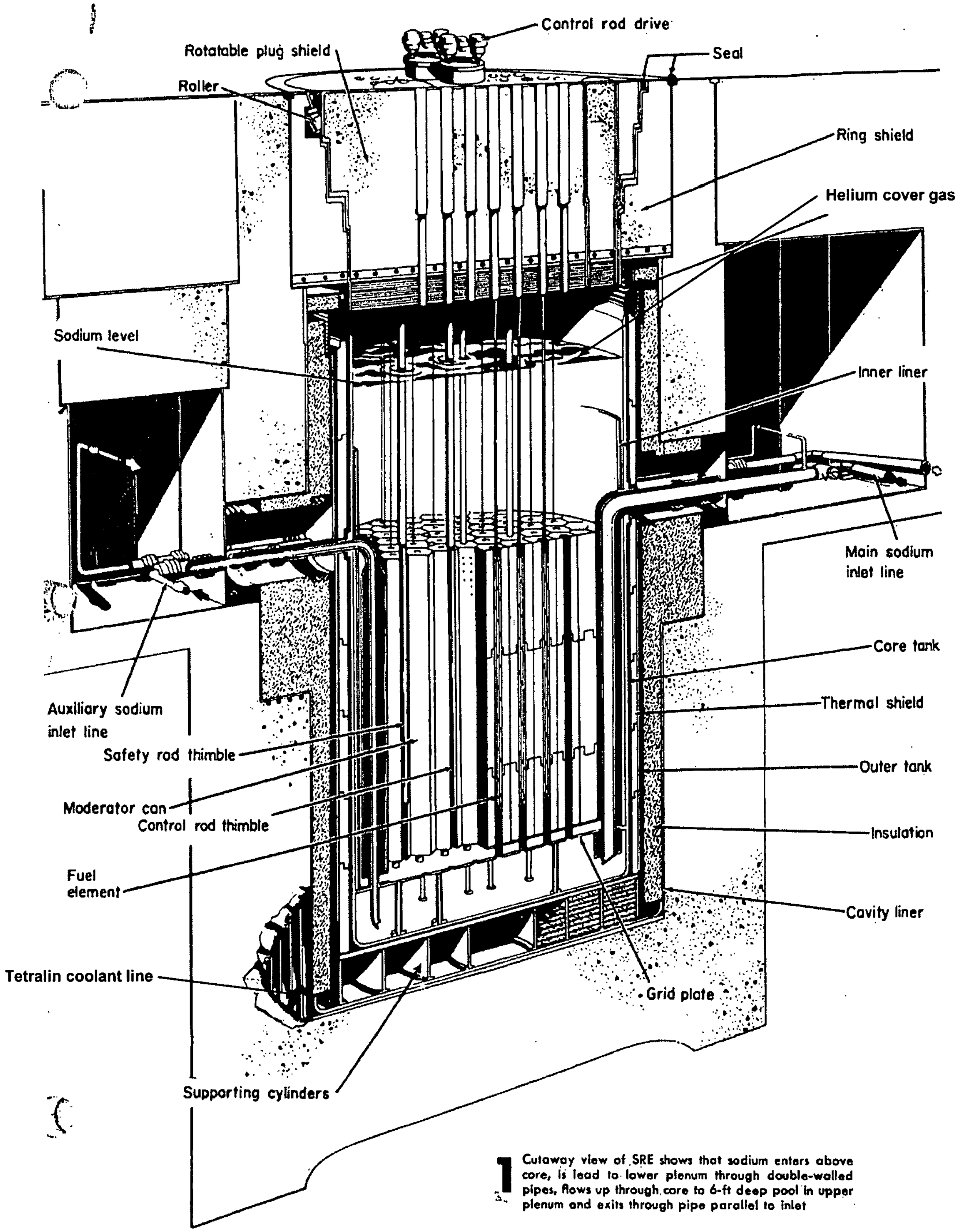
The high bay air was exhausted by means of two fans mounted on the roof, each of which moved 12,000 cfm.[11, p. VI-59] This resulted in 4.8 air changes per hour.

Operation of the SRE could generate gaseous radioactive wastes. Vent lines were connected to the systems containing potentially radioactive gases. These gases were normally discharged from the building vent line to a stack on the roof of the reactor building. Gases were continuously monitored, and if activity rose above a preset level, the gases were automatically diverted to shielded storage tanks until the activity level decayed to a sufficiently low level so that they could be released at a controlled rate, diluted, and discharged out the building stack. The maximum rate of discharge from the storage tanks to the stack was 2.4 cfm and the stack fan mixed this with 25,000 cfm of air, resulting in a dilution of more than 10,000 times.[12]

---

[*] Psig is pounds per square inch gauge and is the amount by which the pressure exceeds the external pressure. Nominally, ambient external pressure is 14.7 psi at sea level. Psia is pounds per square inch absolute, which is the gauge pressure plus external pressure.

[†] For the purposes of accident analysis, the assumed maximum conceivable leak rate from the cover gas into the high bay area was 0.25 cfm (7.1 L/min)[11, pp. VI-55 and B-2], which corresponds to 0.077% of the cover gas inventory per minute. The maximum time assumed for the leak before corrective action would be taken was 2 hours. In a real situation, the leak rate would likely be significantly less.



1 Cutaway view of SRE shows that sodium enters above core, is lead to lower plenum through double-walled pipes, flows up through core to 6-ft deep pool in upper plenum and exits through pipe parallel to inlet

Figure 2. SRE reactor showing fuel elements, sodium pool, and helium cover gas.[2]

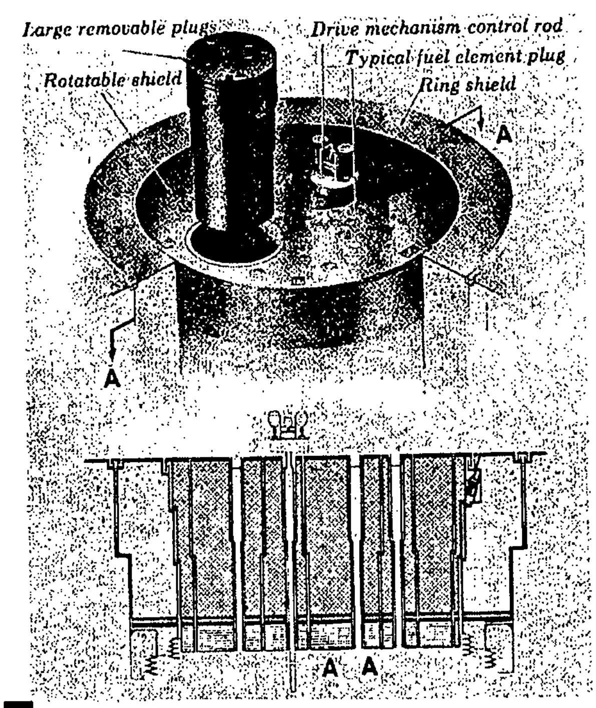


Fig. 24—Detail of the top shield which provides the radiation shield above the reactor as well as the means for access.

Figure 3. Shield above reactor core, sodium pool, and cover gas.[5]

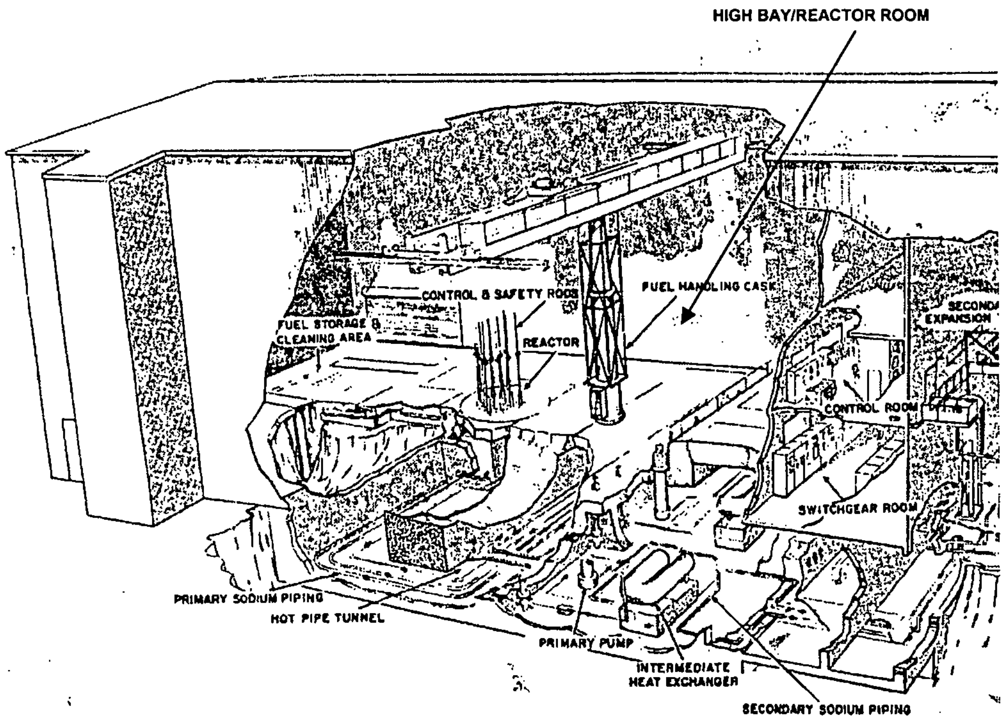


Fig. 3. Sodium Graphite Nuclear Power Plant

Figure 4. SRE reactor facility showing reactor room/high bay above reactor core.[3]

Compressors and controls were located in a shielded vault some distance from the reactor building. Radioactivity in gases normally stemmed from sodium vapor (which contained radioactive isotopes produced as a result of exposure of the sodium to neutrons) and impurities in cover gases exposed to a neutron field.

There were four shielded gas decay storage tanks.[1, p. 2; 6, p. II-A-22; 13] Reference [14, p. BNA03968180] describes the tanks as having a capacity of 350 cubic feet (9,910 L) each at 100 psig. A diagram of this gaseous waste disposal system is given in Figure 5.

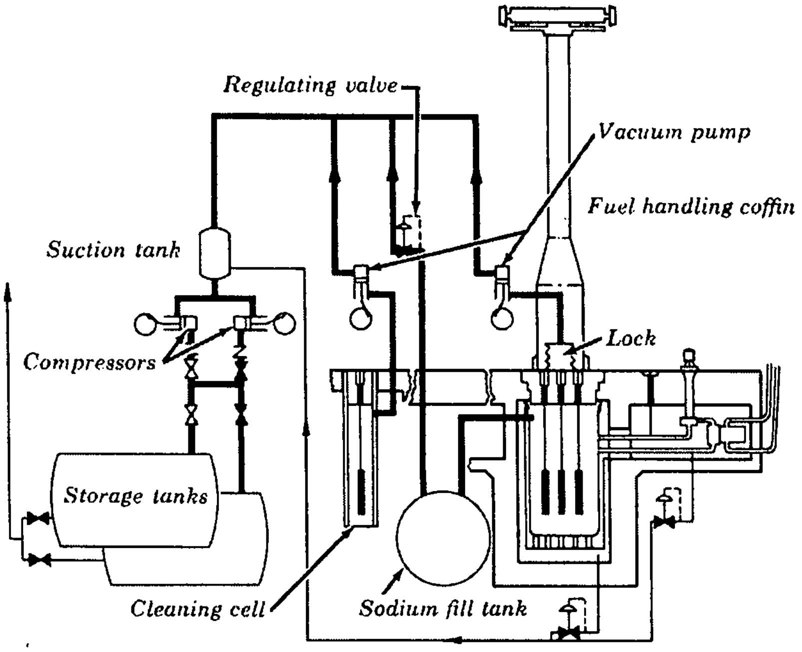


Fig. 74—Diagram of the gaseous waste disposal system. (SRE Photo 74)

Figure 5. Gaseous waste disposal system showing diversion to pressurized storage tanks.[5]

Auxiliary cooling of components of the reactor was provided by a recirculating organic liquid, tetralin. Principal components serviced such as freeze seals, cold traps, etc. are shown in Figure 6. Tetralin is a derivative of naphthalene. Naphthalene consists of two hexagonal "rings" of carbon atoms, with each outside carbon connected to a hydrogen atom; the carbon-carbon bonds are called unsaturated. In tetralin, the carbon-carbon bonds in one of the rings is saturated and each of the carbons is connected to two hydrogen atoms. Each carbon atom has four bonds and each hydrogen has one bond. A drawing of tetralin is shown in Figure 7.

Tetralin is important here because a leak into the sodium caused it to partially dissociate and to react with sodium and create carbonaceous particles that restricted flow of sodium among the fuel channels.

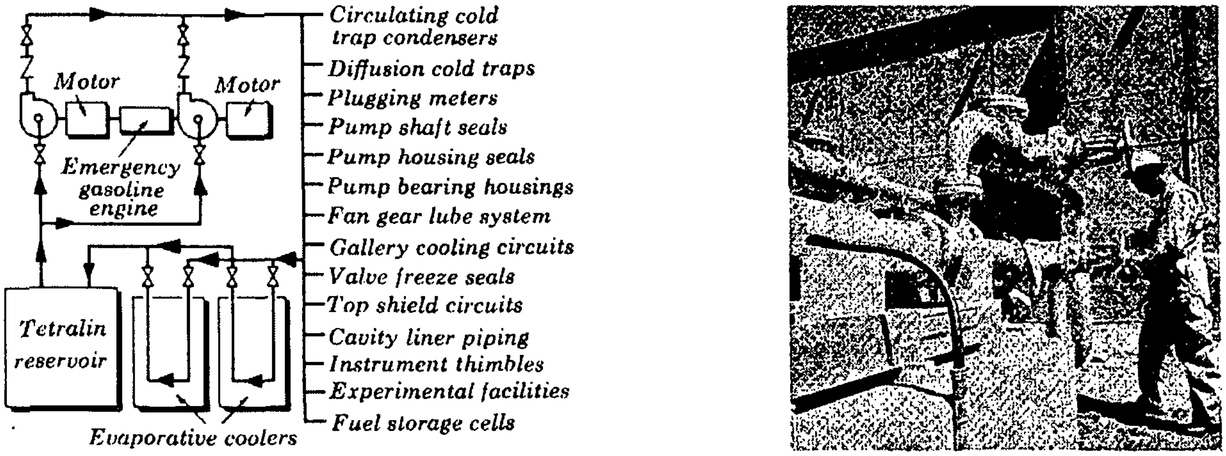


Fig. 59—Diagram of tetralin cooling system showing the principal components serviced. (SRE Photo 59)

Fig. 60—The tetralin pump station. (SRE Photo 60)

Figure 6. Tetralin cooling system with tabulation of components cooled.[5]

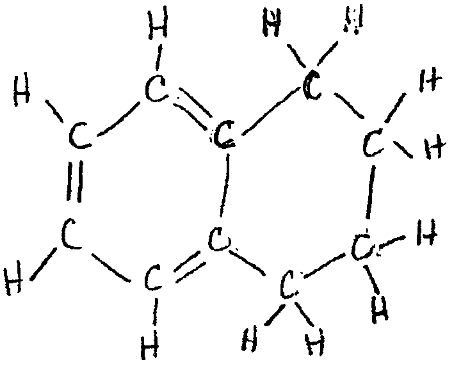


Figure 7. Tetralin molecule, 1,2,3,4-tetrahydronaphthalene, $C_{10}H_{12}$, melting point -35.8°C, boiling point 207.6°C. Used for cooling reactor components.

III. BRIEF LESSON ON VAPORIZATION OF CONDENSED SPECIES

Some of discussion in the following talks about vapor atoms or molecules at equilibrium with the condensed liquid. For example, sodium vapor atoms exist in a bubble of boiling sodium that is in contact with the liquid sodium. Similarly, sodium vapor atoms exist above the hot liquid sodium pool in the SRE reactor. The calculation of concentration of these vapor atoms relative to the modeled concentration of hypothesized iodine molecules (I_2) becomes important in some of the analysis. In order to understand the concepts and analyses a little easier, a brief discussion of vaporization processes is given here.

It is not necessary for a liquid to boil for vapor atoms or molecules of that liquid to exist in a closed region in contact with the liquid. The molecules of a liquid are always in motion, which results in some escaping to the space outside the liquid interface with another gas or in vacuum. Some of the gas molecules subsequently collide in random directions with other gas molecules and return to the liquid. Equilibrium is established when the rate of escape equals the rate of return. If the system is open and the gases above the liquid can escape, evaporation will continue. That is why a glass of water on the counter top will evaporate in a few days, even though it is not at the boiling temperature.

Returning to the concept of a closed system, the concentration of gas molecules in the vapor space in equilibrium with the liquid is a function of the material and the temperature. The associated gas pressure at a given temperature is called the saturation vapor pressure of the substance. When the temperature is increased to the point where the vapor pressure is equal to the external ambient atmospheric pressure, the liquid boils and the liquid temperature will increase no more unless the ambient pressure is physically increased. When the ambient pressure is decreased, the boiling temperature will decrease. Thus, in Idaho Falls where I live, which is at an elevation of 4,730 feet, the atmospheric pressure is 12.3 psi (0.838 atmosphere) instead of the normal 14.7 psi (1 atmosphere) pressure at sea level, and water boils at 203°F (95.1°C) as opposed to 212°F (100°C) at sea level. Thus, my wife has to adjust some recipes to accommodate the high altitude.

The vapor concentration at equilibrium (whether boiling or not) is calculated from the ideal gas law

$$pV = nRT$$

where n in the number of g-mols of vapor species (related to number of atoms or molecules) in a volume of V at a temperature T. T is expressed as absolute temperature. R is the universal gas constant, 0.08206 L-atm/g-mol-deg in metric units. In metric units, T is °C (degrees centigrade) + 273.15, and V is in liters. The vapor pressure, p, is in atmospheres (1 atmosphere = 14.7 psi).

The gas molecule concentration, c, is the number of g-mols per liter, or n/V. Thus,

$$c=\frac{n}{V}=\frac{p}{RT}$$

a simple calculation when the vapor pressure is known. Tabulations are available of vapor pressures as functions of temperature for most species.

When a fission product species such as elemental iodine, I_2, (very volatile) or uranium triode salt, UI_3, (extremely low volatility until very high temperature) is released at high temperature from a fuel, its concentration in a flowing gas will be much less than that corresponding to saturation, that is, less than the vapor pressure of the substance itself, because the release rate is inadequate to saturate the moving gas stream since the quantities in the fuel are so small on a mass basis. It will remain a gas in the flowing stream until the temperature of the gas has been reduced to a temperature corresponding to the saturated vapor pressure associated with the actual gas molecule concentration in the flowing gas stream. Thus, one can sometimes deduce information about the molecular species of the released gas from the observed deposition temperature in association with the gas flow. This is talked about later when experiments involving release of iodine from uranium metal fuels are discussed.

A characteristic of gas mixtures is that the pressure of individual species molecules in the mixture is related to the total external pressure times the ratio of its molecular concentration in the gas to the sum of molecular concentrations of all species in the mixture, that is, its mole fraction. This is called its partial pressure in the mixture. This relationship is called Dalton's law of partial pressures. Thus, if, for example, krypton and xenon atoms are in a sodium vapor bubble in boiling sodium when the external pressure is 1 atmosphere, the total pressure of all the gases in the bubble will be 1 atmosphere. Thus, the partial pressure of sodium vapor atoms will be less than 1 atmosphere. This means that the boiling temperature will be decreased to that value corresponding to the boiling point of pure sodium at the pressure equal to the partial pressure of sodium in the mixture.

It is pertinent, also, to mention here that the elemental form of iodine, I_2, is very volatile and would exist as a gas at nuclear reactor operating temperature. But, salts of iodine, such as uranium triiodide, UI_3, or cesium iodide, CsI, are not very volatile until extremely high temperatures. They are similar to table salt, sodium chloride, NaCl.

IV. HOW ARE FISSION PRODUCTS FORMED?

An overview is given here of pertinent concepts and facts here before getting into details that are described and discussed in subsequent sections.

A nuclear reactor produces energy when a uranium-235 atom is struck by a neutron, which results in the uranium nucleus splitting into two, sometimes three, lighter elements, a process called fissioning. When this happens, a very small amount of the initial nuclear mass is lost and is transformed into energy according to the famous Einstein relativity equation $E = mc^2$. This is manifested in higher binding energies in the medium weight nuclei. The production of 15,000 kWh of electrical energy, which would run a household, including electrical heat, for a year, corresponds to the transformation of 0.0018 g of mass to produce 45,000 kWh of thermal energy at the power plant.

The uranium-235 nucleus, U-235, is made up of 92 positively charged protons and 143 neutral neutrons, the sum giving its mass of 235 atomic mass units. When it is split, the same numbers still remain, but they are now present in pairs of lighter atoms (less an average of 2.5 extra neutrons that are released to continue the fissioning chain reaction process). While a very small amount of the mass is converted to energy, the summed masses are very nearly identical to what was in the original U-235 atom. Each individual U-235 atom fissioned may produce different pairs of fission products that comply with the mass/charge balance constraint. On average, when a very large number of U-235 atoms are fissioned, a distribution of fission products is produced according to Figure 8.

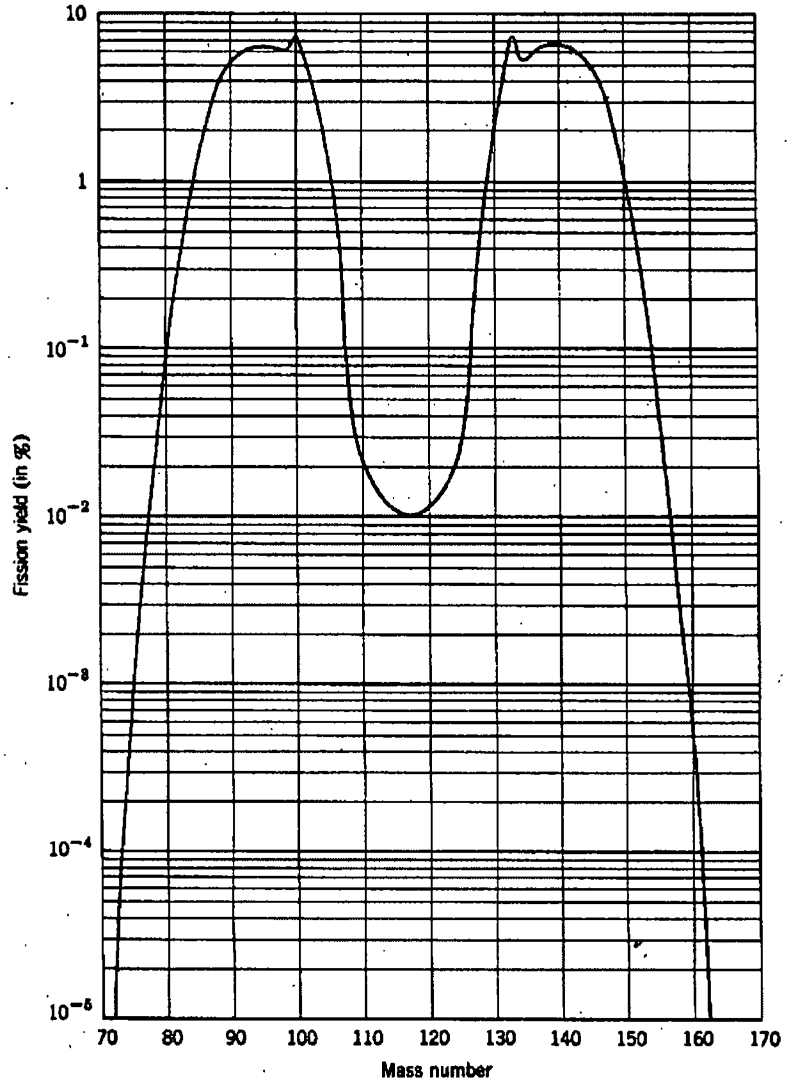


Fig. 10-7 Yields of fission-product chains as a function of mass number for the slow-neutron fission of U^{235}.

Figure 8. Fission yield curve for uranium-235.[15]

What this depicts is the relative numbers of individual fission products produced as a function of how heavy they are, that is, the sum of protons and neutrons in the nucleus. The number of protons defines what the element is. For example, I-131 has 53 protons and 78 neutrons, with a mass number of 131. Cesium-137, another high yield fission product, has 55 protons and 82 neutrons, with a mass number of 137. The reader will note that each of these two fission product isotopes is at a point on the fission yield curve that is near one of the peaks of the curve. The minimum in the symmetrical curve corresponds to one-half the mass of the original U-235 atom. Because the uranium nucleus generally splits into two elements when it fissions, the sum of all the fission product percentages produced, as depicted on the curve in Figure 8, is 200% of the original U-235.

The other thing that is obvious is that not all the fission products are produced each time that a U-235 nucleus fissions. There are some 460 individual isotopes of the approximately 40 elements produced. The combined masses, if formed all at once, would far exceed the original nuclear mass from which they originated. Rather, what happens is an individual U-235 nucleus splits into a symmetrical pair or occasionally three fission product isotopes whose combined sums of protons and neutrons (and the average 2.5 extra neutrons emitted) maintain the mass and charge balances from the original U-235 nucleus. Each atom randomly produces different combinations. The statistical average when a large number of U-235 atoms fission, say, a billion-billion atoms (which would be only 0.0004 gram), results in the distribution given in Figure 8.

Not all of the 460 fission product isotopes in fuel in the reactor are produced directly, but some result from decay of parent isotopes that have short half lives. (Half life is the time that it takes for one-half of the quantity of an isotope to decay to a different species.) For example, only 0.14% of I-131 is directly produced by fissioning of uranium. The balance of 99.86% results from decay of other fission products that mostly have half lives ranging from 39 seconds to 25 minutes: tin-131, antimony-131, and tellurium-131. Each is produced independently, but they also decay sequentially from the first to the next until I-131 results.

When a fission product forms, it is initially in the state of individual atoms in the uranium matrix. These are generally not chemically stable except for a very few elements. The chemical form of a fission product depends on two things. The first is what does it want to become in the given environment? That is, what elements would it like to combine with (including, possibly, itself)? This is determined by chemical thermodynamics and can be accurately calculated. Some, such as krypton, xenon, and palladium, are chemically inert and remain as the elements. The second is, what is, in fact the chemical environment of an atom? What is it near to? It may favorably combine with a number of different other elements, in a hierarchy of preference, and, when near an abundance of lower preference atoms, may combine with them. In the case of low burnup fuels, the most immediate element in abundance is uranium. At the maximum burnup of the SRE fuel of 0.1 percent, for example, there would be more than 550 atoms of uranium around each fission product atom, more than 550, because not all fissioned uranium atoms form that one fission product. Looking at the fission yield curve above, I-131 has a fission yield of about 3%, so each atom would be surrounded by about 15,000 uranium atoms if I-131 did not decay. Actually, because I-131 decays with a half life of 8 days, the so-called steady state concentration reached as previously formed atoms decay while new ones are being made is much less relative to the number of uranium atoms. For the SRE fuel, the uranium-to-iodine atom ratio was about 280,000-to-1 at the time of Run 14 on July 13, 1959, see Table II in Section VI.

When thermodynamic calculations are done for the uranium metal fuel, the results predict that iodine would form cesium iodide, CsI (see Section VI). But, experiments to determine the form of iodine released from melted metal fuels indicate the likely presence of uranium iodide, UI_3. One can see why this might be if the iodine reacts with uranium before it gets a chance to find a cesium atom by diffusion through the matrix. If, in the thermodynamic calculations, the CsI (and other fission product metal iodides that would also form ahead of UI_3 in the absence of cesium) are eliminated as possible products, then the prediction is for the formation of UI_3. Thus, experimental evidence is found of UI_3 and, perhaps, some CsI in irradiated uranium metal fuels. Since there are about 15 to 30 times more cesium atoms formed as iodine (the relative amounts depend on fuel burnup, power level, length of time after shutting down reactor, etc.) even when some CsI is formed, the bulk of cesium remains as the unreacted element.

The thermodynamic calculations do not predict the formation of elemental iodine, I_2, and experiments bear that out. If, for the moment, one does assume that it should be a stable species (though it is not), it would be usurped by the relatively massive amount of uranium grabbing the iodine before two iodine atoms could find each other. But, the underlying chemical fact is that elemental iodine, I_2, would not form even if two atoms found one another or if two UI_3 molecules interacted.

There is a difference in the chemistry of fission products in uranium oxide fuels compared with uranium metal fuels. In oxide fuels that are used in modern commercial power reactors, the uranium oxide does not chemically want to shed its oxygen for the iodine. It will combine with cesium to form cesium uranate, such as Cs_2UO_4, $Cs_2U_2O_7$, or $Cs_2U_4O_{12}$. This also ties up the most of the cesium so that it cannot react with the iodine. Some cesium may also combine with molybdenum to form a binary oxide, cesium molybdate. Other fission products that could otherwise combine with iodine are, similarly tied up: barium, which is an oxide precipitate like cesium; strontium, lanthanum, cerium, and neodymium dissolved as oxides in the uranium oxide fuel matrix (see Section V). Thus, for oxide fuels, in general, most iodine in the oxide fuel matrix forms I_2. While the I_2 generally remains in fuel during normal operations it would be released as a volatile gas from melted oxide fuels.

V. URANIUM OXIDE FUELS AND METAL FUELS ARE DIFFERENT WITH RESPECT TO FISSION PRODUCT CHEMISTRY AND BEHAVIOR

The behavior of fission products during a nuclear reactor incident in which fuels are damaged depends importantly on the chemical form of the uranium from which the fuel was fabricated. Today's commercial nuclear power plants in the U.S. are light water reactors where the fuel is in the form of uranium dioxide, UO_2, that is cooled with water. By contrast, (significant for this case) the SRE was fueled with assemblies of stainless steel-clad uranium metal that were cooled with flowing liquid sodium metal. (A few elements had uranium metal alloys in which uranium was combined with other metals. None of the failed elements contained oxide fuel.) Studies of postulated reactor accidents usually have investigated what happens to built up fission products in the fuels if they are melted. In addition to significantly different chemistry, an important difference is also that uranium dioxide melts at 5212°F (2878°C), vs. 2069°F (1132°C) for uranium metal. The differences in chemistry and melting temperature lead to substantial fission product volatilization from oxide fuels in melting accidents, in particular, iodine, that does not occur from metal fuels.

The fact that the SRE reactor used uranium metal fuels instead of the oxide fuels that are used in conventional commercial power light water reactors is a very important aspect regarding the consequences of the SRE fuel cladding failures July 1959. The SRE uranium metal fuel itself did not melt. The relevant fission product chemistry is different between the SRE metal fuel and oxide fuels, especially with respect to iodine. As will be shown in substantial detail here and in subsequent sections, the fission product iodine, which Dr. Makhijani claims was released from SRE, did not form as volatile elemental I_2 in the metal fuels of the SRE, but as an iodide salt with other fuel constituents. That fact, along with the fact (discussed separately in Section IX.A) that the low burnup (small amount of irradiation and, thus, fissioning) of the metal fuels did not produce enough gaseous fission products of any sort (including krypton and xenon) to form bubbles that could migrate out of the fuels, resulted in the iodine being retained in the uranium metal and alloy fuel elements when the cladding failed, except for the very small amount that was released similarly to other non-volatile fission products into the sodium.

In metal fuels iodine exists as the salt uranium triiodide (UI_3) or cesium iodide (CsI). Such is not the case in oxide fuels. Thus, both iodine and cesium fission products behave differently in heated oxide fuels. This is because oxygen forms binary compounds between uranium and cesium and between molybdenum (a fission product) and cesium. At the same time the bulk of uranium is present as uranium oxide. Thus, in oxide fuels neither uranium nor cesium is completely available to form iodides and much of the iodine exists as elemental iodine, I_2.

In 1985, Kleykamp published a seminal paper on the chemical state of fission products in oxide fuels, based on both theoretical and micrometer scale experimental elemental analysis studies.[16] He found that the chemical state of the fission products can be classified into four groups:

(1) fission gases and other volatile fission products: Kr, Xe, Br, I;
(2) fission products forming metallic precipitates: Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Te;
(3) fission products forming oxide precipitates: Rb, Cs, Ba, Zr, Nb, Mo, Te;
(4) fission products dissolved as oxides in the fuel matrix: Sr, Zr, Nb, and the rare earths Y, La, Ce, Pr, Nd, Pm, Sm.

As is seen, molybdenum, Mo, can exist both as metallic precipitate and as an oxide. There are continuous transitions between the groups (2) and (3) due to similar oxygen potentials of some fission product oxides and the fuel, which changes its composition during the fission process. Furthermore, the halogens of group (1) (Br, bromine, and I, iodine) can also form compounds with the most electropositive fission products, e.g., cesium, Cs. Thus, the situation regarding cesium and iodine is not well defined, but it is clear that elemental iodine (I_2) can be formed at the high oxidation potential of irradiated uranium oxide fuels.

The quantitative effect of this depends on the amount of burnup of the oxide fuel, which is related to the amount of fissioning that has occurred and resulted in formation of fission products. In oxide fuels, the uranium starts out as the chemical form UO_2. But as uranium fissions and is depleted, the oxygen-uranium ratio increases and the oxygen potential of the system increases, which pulls cesium away from iodine. Uranium oxide reacts with cesium or cesium iodide to form cesium uranates, such as Cs_2UO_4, $Cs_2U_2O_7$, or $Cs_2U_4O_{12}$. Furthermore, at high oxygen potentials, fission product molybdenum metal can oxidize to also react with cesium, forming Cs_2MoO_4.[16, 17] that strips the cesium from cesium iodide that normally suppresses iodine volatility at high temperatures of a reactor accident.

In an oxide fuel melting accident, elemental iodine, I_2, can be released. These considerations do not apply to metal fuels of the kind used in the SRE in which iodine exists as iodide in the form of UI_3 or CsI. Furthermore, other circumstances that prevented the release of iodine from the SRE were the fact that the uranium metal fuels inside the cladding never melted (except for the very small fraction that alloyed with the cladding at a lower temperature, see next paragraph and Section VII), and the burnup was not high enough to yield enough fission products to cause gaseous fission products to form large bubbles that could migrate out of the fuel (see Section IX.A).

A digression at this point is in order re: alloying of uranium metal with cladding. An explanation of alloying between metals and formation of a eutectic composition that can melt below the melting temperature of either of the pure metals is in order to understand the mechanism of the SRE cladding failure that is discussed later in Section VII. Uranium metal melts at 2,075°F. Iron melts at 2,795°F. Stainless steel that contains iron alloyed with other constituents melts at 2,624°F. But, uranium and iron can form an intermetallic compound called an alloy whose melting point is lower than any of these temperatures. It is analogous to the antifreeze mixed with water in an automobile's radiator. Water freezes at 32°F. Antifreeze, which is ethylene glycol, freezes at 11°F. But when ethylene glycol, which is soluble in water, is mixed with water at a 50:50 volume ratio, the freezing point of this mixture is -34°F. As the ethylene glycol ratio increases up to about a 60:40 volume ratio, the freezing (melting) temperature decreases further to -58°F. Beyond that, the freezing/melting temperature begins to increase again. The composition of the minimum freezing temperature is called the eutectic composition. Eutectic means a mixture of two or more substances that has the lowest melting point. If a 60/40 mix of ethylene glycol and water is frozen at a temperature below -58°F and then warmed, it will melt at -58°F. A figure that plots the freeze/melt temperature versus the composition is called a phase diagram.

Another familiar system is a mixture of salt (sodium chloride) and water used in a home ice cream maker. When an excess of salt is added to water and ice, the eutectic mixture forms and the freezing temperature is lowered to -6.2°F so that the cream can be frozen.

This identical situation occurs between uranium and iron. The phase diagram for the two is shown in Figure 9. The eutectic point at 725°C (1,337°F) is indicated. The temperatures indicated in the diagram are centigrade. Thus, when uranium and iron metals are in contact and heated, the iron and uranium can diffuse into each other and form a melt in that region at 1,337°F, a process called alloying. The consequence of this is discussed in Section VII.

Discussions of actual release characteristics of iodine from heated and melted metal fuels and of iodine behavior in liquid sodium are provided in Sections VIII through X. Experimental data on the chemical form of iodine released from heated metal fuels are summarized here. Castleman determined the deposition temperatures of fission products released from both U metal and U-3.5% Mo metal alloy irradiated fuel when melted at 2282°F (1250°C).[18, 19] The released iodine deposited at 536°F (280°C), well above the temperature that would correspond to saturated I_2, that is, the temperature at which I_2 vapor would condense. When I_2 vapor was passed over uranium metal, a deposit of a uranium-iodide compound formed at the same temperature. When the irradiated fuels were heated in the presence of air where oxygen reacted with the fuel, the released iodine deposited below 100°C, indicating conversion to elemental I_2. This is consistent with the chemistry in oxide fuels versus metal fuels as discussed above.

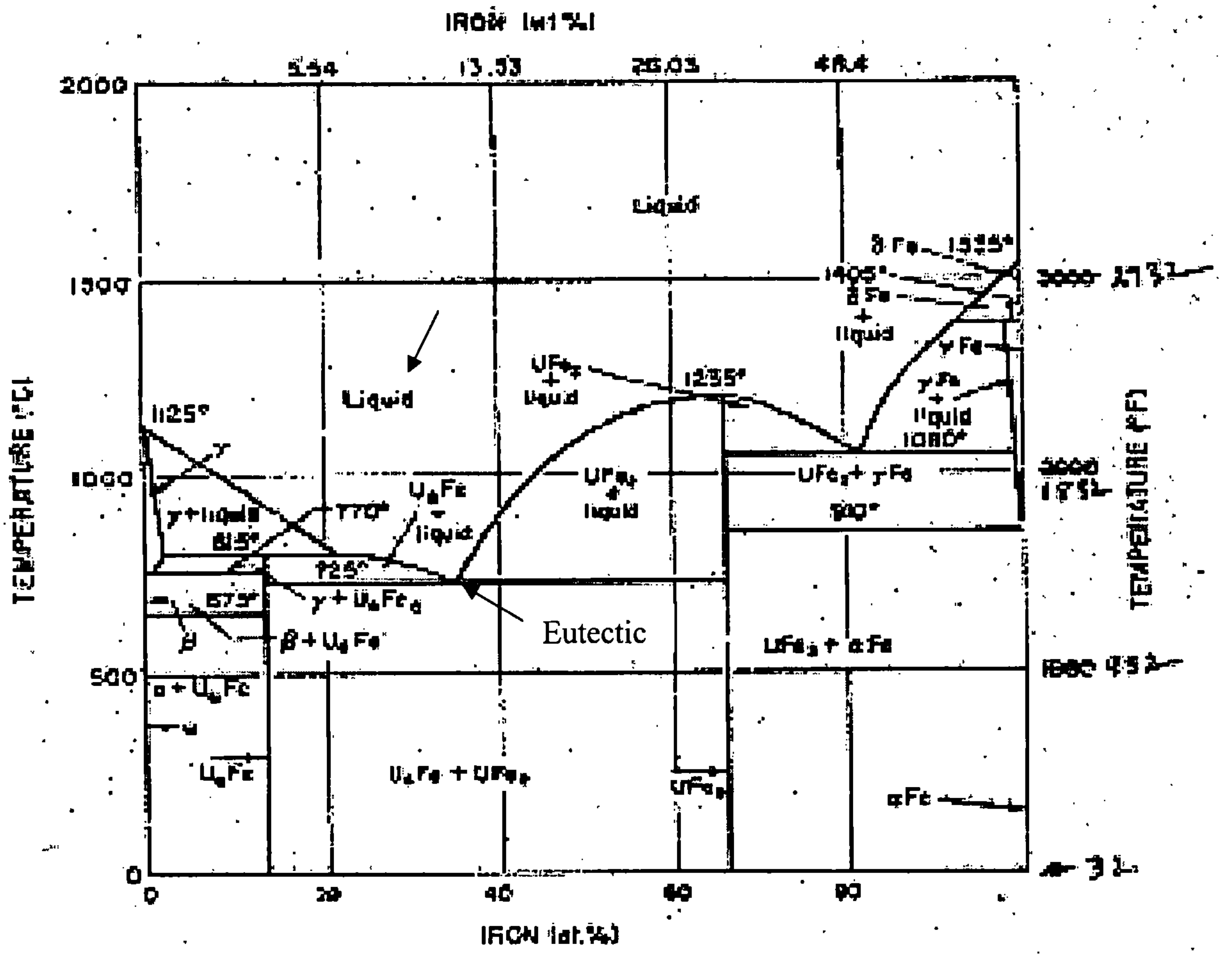


Figure 15. Constitutional Diagram of Uranium-Iron System

Figure 9. Uranium-iron phase diagram showing the eutectic point.[20, p. 30]

Cesium and other fission products deposited at different temperatures than iodine, indicating the absence of cesium iodide (CsI) and other fission product iodides. Parker and Barton comment on this: "Since fission-produced iodine atoms are surrounded by uranium atoms, it appears that favorable conditions for reaction exist."[21, p. 541]

These experiments demonstrate that elemental I_2 is not released from metal fuels like were used in the SRE.

VI. THERMODYNAMIC MODELING SHOWS THAT IODINE FORMS METAL IODIDES IN THE URANIUM METAL FUEL

Theoretical calculations can be performed to predict the chemical species present in irradiated uranium fuel. This is done using chemical thermodynamics, which calculates whether or not a species can form from the initial composition of elements present at specified quantities. Results can be compared with experimental data to provide a theoretical basis for the observations.

Thermodynamic calculations are based on using a database of thermodynamic properties of individual species that have been experimentally determined for them. The specific data used are called standard enthalpy of formation at 25°C, standard entropy at 25°C, and standard heat capacity expressed as a function of temperature. Using these, one can calculate the standard free energy of formation of each species at the temperature of interest. When the system is at equilibrium, that is, the elements are in the combination of compounds that is most stable, the total free energy of the system is zero.

The fission product and uranium composition of the SRE fuel at the time of the cladding failures was used as an input to commercial software that automatically performs the calculation of the equilibrium composition. The software used is called HSC Chemistry for Windows, version 5.1.[22] It has a database for more than 17,000 species. The equilibrium composition of a system is calculated by inputting the starting composition and the set of potential compounds that could form to evaluate as possible equilibrium species.

The input composition for the thirteen failed fuel elements of SRE was provided by Daniel & Associates[8] using the ORIGEN II code that is standard in the nuclear industry for calculating fission product and actinide formation in irradiated fuels. The quantities of individual isotopes of each element of interest were calculated and summed for the element to obtain its total mass and number of atoms (which is needed for the thermodynamic calculation). The number of atoms was converted to g-atoms[*] for input to the HSC program. The elements selected and methodology were in part based on results of similar calculations for uranium-aluminum alloy fuels performed by Adams et al.[23, pp. 17-22]

The quantities of the major components were calculated and inputted to HSC. They are given in Table II. Except for uranium, all are fission products that were produced during irradiation of the uranium.

The thermodynamic modeling results for the equilibrium composition of the fuel are summarized in Table III.

---

[*] The number of g-atoms (gram-atoms) is related to the number of atoms and is the quantity expressed as the atomic weight of the element in grams. It is calculated by dividing the number of atoms by Avogadro's number, 6.023×10^{23}, which is the number of atoms in a quantity of the molecular weight of the element expressed in grams. When one is considering molecules, the equivalent term is g-mols.

Table II. Uranium and Fission Product Quantities in SRE Failed Fuel Elements on July 13, 1959.

Species		
Name	Chemical Symbol	Quantity, G-Atoms
Antimony	Sb	0.0009
Barium	Ba	0.1933
Bromine	Br	0.0050
Cerium	Ce	0.4389
Cesium	Cs	0.4109
Iodine	I	0.0134
Lanthanum	La	0.8049
Krypton	Kr	0.4061
Molybdenum	Mo	0.4841
Neodymium	Nd	0.8090
Niobium	Nb	0.1586
Ruthenium	Ru	0.3639
Strontium	Sr	0.2172
Technetium	Tc	0.7274
Tellurium	Te	0.0806
Uranium	U	3787.86
Xenon	Xe	0.6120
Zirconium	Zr	0.7293

Table III. Thermodynamic Modeling Result for SRE Fuel Equilibrium Composition.

	Equilibrium Quantity	
Species	G-Mols	% of Element Input
U	3787.77	99.9977
Zr	0.7293	100
Xe	0.6120	100
Mo	0.4841	100
Nd	0.8090	100
Cs	0.2313	56.29
Ce	0.4389	100
Sr	0.2172	100
Ba	0.1933	100
La	0.8040	99.89
Tc	0.7274	100
URu_3	0.1213	100 Ru; 0.0032U
Kr	0.4061	100
Cs_2Te	0.0806	39.23 Cs; 100 Te
Nb	0.1586	100
CsI	0.0134	3.26 Cs; 100 I
CsBr	0.0050	1.22 Cs; 100 Br
LaSb	0.0009	0.11 La; 100 Sb

The results show that the halogens, iodine and bromine, would be completely converted to the cesium salts. However, at the extremely small concentrations of the fission products in the uranium matrix, it is likely that interaction with the uranium would occur in the absence of nearby cesium and the uranium halide salt would be formed. This is, in fact, what is observed in experiments and from other metal-fueled reactor fuel failures that are directly related to events that occurred in SRE – see Sections V, VIII, IX, and X. The relevance is that the fuels in SRE were also metal uranium fuels that would have similar chemistry to the experimentally observed chemistry in these other situations.

It turns out that other fission product metals will form iodides preferentially to uranium, as well, if cesium is eliminated as a reactant. When the thermodynamic modeling is done by sequentially eliminating the iodides of the different fission product metals, it is found that the following order of forming iodides occurs: cesium iodide (CsI), barium iodide (BaI_2), strontium iodide (SrI_2), lanthanum iodide (LaI_3), cerium iodide (CeI_3), neodymium iodide (NdI_3), and, finally, uranium iodide (UI_3). In no instance of eliminating these iodides does elemental iodine, I_2, form.

The specific mix of iodides that forms depends on a number of factors that aren't related to equilibrium. Equilibrium occurs when sufficient time has passed to allow the overcoming of a number of potential barriers, such as diffusion in the uranium matrix, for specific interactions to occur. The physical situation is such that UI_3 is formed, but, if not, cesium iodide is the next possibility, and it has been observed in some tests. As UI_3 diffuses out of the uranium matrix, some can interact with cesium atoms to convert to CsI and U.

It may be noted that, if slightly different fuel compositions are inputted into the HSC program from inventories of different dates or from alternative source term calculations by others, the resulting calculated equilibrium chemical composition in terms of what form each element is in will not be changed, other than the fraction of free cesium that is left after 100% of the I, Br, and Te are combined with it and fraction of free La after 100% of Sb is combined with it.

These theoretical calculations corroborate experimental observations of the chemical form of iodine released from metal fuels being uranium iodide and/or cesium iodide salt and not elemental I_2.

VII. MECHANISM OF FUEL FAILURE IN SRE SHOWS THAT THE URANIUM FUEL DID NOT MELT

The fuel cladding failures occurred as a result of a tetralin leak into the sodium. The tetralin decomposed in the sodium at a temperature of about 675°F (357°C) to carbon and a mixture of low molecular weight aliphatic hydrocarbons.[6, p. IV-B-1] The quantity of carbon formed exceeded its solubility in sodium and the carbonaceous particulate material caused partial restriction of sodium flow in some fuel channels. By approximately July 12, 1959, the resultant decrease in cooling led to some of the fuel elements increasing in temperature.

Three contributing mechanisms led to cladding failures and ruptures – all related to the high temperatures resulting from the reduced sodium flow. The first is that the increased temperature with uranium fuel slugs in contact with the stainless steel cladding caused rapid diffusion of uranium into and alloying with the stainless steel. At locations where the temperature exceeded the melting point of the iron-uranium eutectic, 1,337°F (725°C), this diffusion resulted in the formation of an alloy with some liquid phase present. It ultimately resulted in failure of the cladding of some of the fuel elements, though the fuel did not melt. (Uranium melts at 2,075°F.) Over the course of the next several days, most likely by about July 23rd cladding of 13 of the 43 elements failed. Of the 13 failed element claddings, three showed no evidence of eutectic melting. However, thermocouples showed that they cycled several times through the α-β (alpha-beta) phase transformation temperature for uranium of 1,225°F (663°C). A 5% anisotropic volume increase results during the change from the orthorhombic α phase to the tetragonal β phase and likely ruptured the cladding from swelling.

Aggravating both of these mechanisms, especially the eutectic-caused cladding rupture, is the high vapor pressure of the NaK bonding liquid metal alloy in the gap between the uranium slugs and the cladding. NaK boils at 1,445°F (785°C), substantially below the boiling point of sodium of 1,621°F (883°C). That is the temperature at which the pressure exerted by the NaK vapors is 1 atmosphere (14.7 psia). As the temperature of the fuel slugs increased above 1,445°F the vapor pressure of the NaK would have increased exponentially above atmospheric pressure and resulted in large internal stresses on the cladding that could contribute to rupture of cladding well before the sodium in contact with the cladding boiled. Any hypothetical gases in the gap would have been released then. Any hypothetical iodine would have been in the form of sodium iodide and potassium iodide from reaction with the NaK.

The consensus of all investigations is that the uranium did not melt but that the claddings failed by either the low temperature eutectic formation and/or by swelling from cycling through the alpha-beta temperature. There was no evidence of uranium melting.[1, p. 20] An evaluation of the mechanism of reactivity changes in the core was based on the assumption of no melting of the uranium slugs.[6, p. IV-D-1] The maximum temperature reached in the fuel was estimated in the interim report by considering the operating parameters and conservative assumptions to be about 1,400°F for a short time. If low heat removal from the fuel extended over a longer period of time, fuel temperatures of 1,500°F to 2,000°F range were possible.[6, p. IV-D-20]

There is considerable importance put on when the majority of the fuel claddings failed and potentially released fission products. In order for Dr. Makhijani's theory for I-131 release to the environment to be scientifically valid, they would have had to fail and release the iodine, krypton, and xenon before July 15, the date that he claims the cover gas contents were vented to the stack. He acknowledges that no releases of the cover gas to the stack occurred after July 15. The evidence for when the failures likely occurred is discussed in Section XIII. An input to that discussion is the temperature history of the fuels and the fuel channel sodium exit temperatures in relation to when the above failure temperatures might have occurred.

Figures IV-D-5a and IV-D-5b in the interim report[6] plot data for the fuel slug temperature for R-67 (not a damaged fuel) and the fuel channel exit temperature of sodium for channel 54 for the entire Run 14, from July 11 to July 26. They track together. The plots show some increase above the baselines from July 12 to 15. The slug temperature rose from 500°F on July 12 to 550°F on July 15 and then decreased until July 20 when it rose rapidly to a maximum of 730°F on July 23-24. Similarly, the fuel channel exit temperature attained 580°F on July 15, decreased and increased rapidly on July 20, reaching a maximum of 790°F on July 22, holding above 700°F through July 14.

The temperature history of the element in channel 55 (a damaged fuel) and the channel sodium exit temperature from July 15 to July 26 are displayed in Figure IV-A-5 of the interim report. It shows fuel temperature of about 450°F on July 15. The chart recorder ran out of paper on July 20, but reached a maximum of 1350°F then (above both the U-Fe eutectic and the alpha-beta transition temperatures). A fresh chart 12 hours later started at about 800°F and rose to 1,450°F peak temperature on July 22. Similarly, the channel exit temperature of sodium ranged from 400°F on July 15 to a maximum of 900°F on July 22-23.

Significantly, the temperature of R-55 from July 23 to shutdown on July 26 (three days) did not decrease back to the level of the July 15 temperature (two to three days of any postulated temperature excursions). This indicates that a temperature excursion between July 12 and 15 was not likely.

A detail of the channel R-55 fuel element temperature for the period July 22-23 is given in Figure III-8 of the final report[24]. It shows about four dozen cycles through the alpha-beta transition temperature from 3:00 p.m. July 22 to 8:00 a.m. July 23. Similarly, Figure III-7 shows the sodium exit temperature of that channel cycling rapidly during that time, reaching a maximum on midnight of July 22, the same time that the fuel temperature reached a maximum. The outlet temperature on channel R-24 (a failed element) (Figure III-5 of the report) showed similar behavior. The outlet temperature for channel R-25 showed cycling until 4:00 p.m., then a steady, rising temperature that peaked from 5:00 p.m. to midnight.

These all point to conditions for substantial fuel damage in the July 22-24 time period and, in fact, that conditions were more likely favorable for failure then than during July 12-15. This is not to say that a few, two or three elements did not fail in the earlier period, but, most likely, most damage occurred later.

Measurements on the failed element R-24 showed that it had formed about 800 grams of the iron-uranium eutectic material that forms at 1337°F/725°C [24, p. V-1]. For the U-Fe eutectic composition of 88.8 wt% U [20, p. 30], this corresponds to 1.05% of the uranium in the element. The evidence is that most of the failed claddings involved eutectic formation. If one assumes that cladding of about 8 assemblies (60%) were alloyed and the same percentage of eutectic formation in all of them, this would correspond to a total of about 0.65% of the total uranium inventory of 13 elements. This provides a rough estimate of the total amount of uranium that may have formed a eutectic melt with the cladding, which was at a temperature substantially below the pure uranium melting point. It is fortuitous and may be of significance that this also corresponds approximately to the percentage of fission products released to the sodium and cover gas (see Table VI in Section XIII).

VIII. DATA ON IODINE BEHAVIOR DURING NORMAL OPERATIONS OF THE EBR-II SODIUM-COOLED REACTOR SHOW THAT IODINE IS NOT RELEASED FROM FAILED-CLADDING METAL FUELS

The Experimental Breeder Reactor-II in Idaho was a metal-fueled, sodium-cooled reactor like the SRE. Experience in EBR-II during its years of operation has shown that iodine was never released from fuels during normal operations at about 720°F (382°C). John Krsul is a recently retired analytical chemist who made many measurements for iodine in sodium and the cover gas of EBR-II.[25] During cladding failures in EBR-II metal fuels, iodine was never measured anywhere but in the fuel. Even during severe failures, any small amount of iodine that came out of the fuel was in the reduced iodide (salt) form, demonstrating that elemental iodine, I_2, isn't released for metal fuel. This is consistent with all other information that indicates iodine is not present in metal fuels as elemental iodine and that it is retained in the fuel during severe accidents in which the uranium metal fuel does not melt.

Iodine-131 was never found in the stack gases during the years of operation of EBR-II. The detection limit was 2000 dps I-131; 0.054 μCi would have been detected. The only time I-131 was detected was when a small release occurred from the analytical laboratory during analysis of a one gram sample of freshly irradiated fuel.[25]

IX. EXPERIMENTS OF HEATING METAL FUELS TO FAILURE SHOW THAT NO GASES ARE RELEASED PRIOR TO MELTING THE METAL MATRIX

Numerous studies have been conducted on metal fuels, like those used in the SRE, to characterize fission gas and other fission product releases from the fuel during heat up and melting. They show demonstrably that fission gases, including krypton and xenon, are not appreciably released before the metal matrix is melted. Further, the evidence is that iodine released at higher temperatures is not released in the elemental form, I_2, but forms metal iodide with the uranium or fission products.

A. Fission Gas Bubble Formation and Release in Metallic Fuels Does Not Occur at the Low Burnup of the SRE Fuels

Metallic fuels are normally characterized by relatively low release of fission gas.[26, p. 200] The mechanism of release of fission gases (i.e., xenon and krypton) from fuels is the formation of gas bubbles that interconnect to form pathways out of the fuel matrix. The process has been thoroughly characterized and the knowledge gained is summarized in a 1999 paper by Argonne National Laboratory researchers, who developed and characterized the performance of metal fuels for the Experimental Breeder Reactor-II[27] and by British researchers for pure unalloyed uranium fuel.[28] The analysis shows that fission gases will not be released from uranium metal fuel until it has swelled to volumes corresponding to burnups of about 1 %. The SRE uranium metal fuels were burned to 0.1% or less.

Fission gas (e.g., xenon and krypton) does not dissolve into metallic uranium, and, so, precipitates as separate gas bubbles. A certain small concentration of gas may be held in dynamic solution in the uranium matrix, as a result of fissioning, but most fission gas will precipitate as small bubbles, some of which then grow by acquiring more gas atoms. At some burnup level, when sufficient fission gas has been generated, the gas bubbles tend to maintain an equilibrium pressure by balancing internal pressure P_i against bubble surface tension γ and external pressure P_{ex} through growth (i.e., increase in bubble radius). This mechanism can lead to high swelling rates at high temperatures.[27, p. 88]

Fission gas release has been correlated with swelling and it appears to be rather independent of fuel alloy.[27, p. 90] In uranium metal, swelling during irradiation normally occurs by three mechanisms in normal operation.[27, p. 88] The first is the production of solid fission products forming twice as many atoms as were fissioned. This results in about 2 to 3% swelling per atom percent burnup and predominates up to about 1% burnup.[29, p. 106]] The second type occurs from anisotropic (radial) swelling of the alpha phase [that exists to 1236°F (669°C)] as a result of temperature increase and occurs at about 932°F (500°C). It is characterized by large, irregular cavities and is called cavitational swelling. Cavitational swelling is not fission-gas driven and, consisting of voids, is compressible. However, at high burnup, fission gas will collect in the preformed cavities.

Swelling in the higher temperature beta and gamma phases occurs by growth of fission gas bubbles, in addition to the swelling from production of solid and liquid fission products. Swelling from accumulated fission-gas bubbles predominates at 1% burnup and higher.[29, p. 106]

Very little fission gases are released from metal fuels such as used in SRE. The relationship between percent release and swelling[27, p. 90; 30, pp. 33-34] is shown in Figure 10. Until swelling approaches a 20 to 30% volume change, very little fission gas is released. Most gas release occurs at about 30% swelling.[27, pp. 89-90] This corresponds, also, to a theoretical model that predicts the interconnection of bubbles should occur after fuel volume increases of 33.3%. The interlinked bubbles provide paths for fission gas release to the surface of the fuel.[31] This occurs at about 3% burnup for the alloy fuels.[32, 26, p. 200] This is also documented elsewhere in the literature. For EBR-II fuels, swelling of 6% to 14% occurs at a burnup of 1%, depending on alloy type.[29, p. 108]

Metal fuels swell anisotropically, that is, primarily in the radial direction. For a 15% change in diameter, the axial growth in alloy fuels is only 2 to 8%.[27, p. 91] The axial growth in unalloyed uranium metal may be a bit greater, but still would not contribute more than about 5% to volume increase, considering that volume is proportional to the square of the diameter. The SRE fuels were scheduled to be changed after Run 14 because the swelling was causing the uranium fuel to approach contact with the cladding. As explained below, this provides an indication that the SRE fuels had not swelled sufficiently to release fission gases. This swelling to the cladding corresponds to a cross sectional area increase of 5.4% at contact with the cladding with a corresponding total volume swelling of about 5.6%. If the diameter swelling prior to changeout was about 90% of the 0.01" gap, the volume increase was near the 5.6% at the start of Run 14, well below what would release gases. This swelling of SRE fuel is indicated by the arrow in Figure 10, which clearly shows it is way below the region of fission gas release. This swelling for SRE fuel is reasonable in the light of the established 30% volume increase for 3% burnup for the alloy fuels. The established linear relationship[6, p. IV-A-5, 33] would correspond to 1 volume percent at 0.1 burnup of SRE fuels. Since uranium metal swells at a greater rate than alloy fuels, three to five times the swelling may have occurred in the SRE fuels, or 3 to 5 percent, reasonably in line with the 5-6 % estimated above.

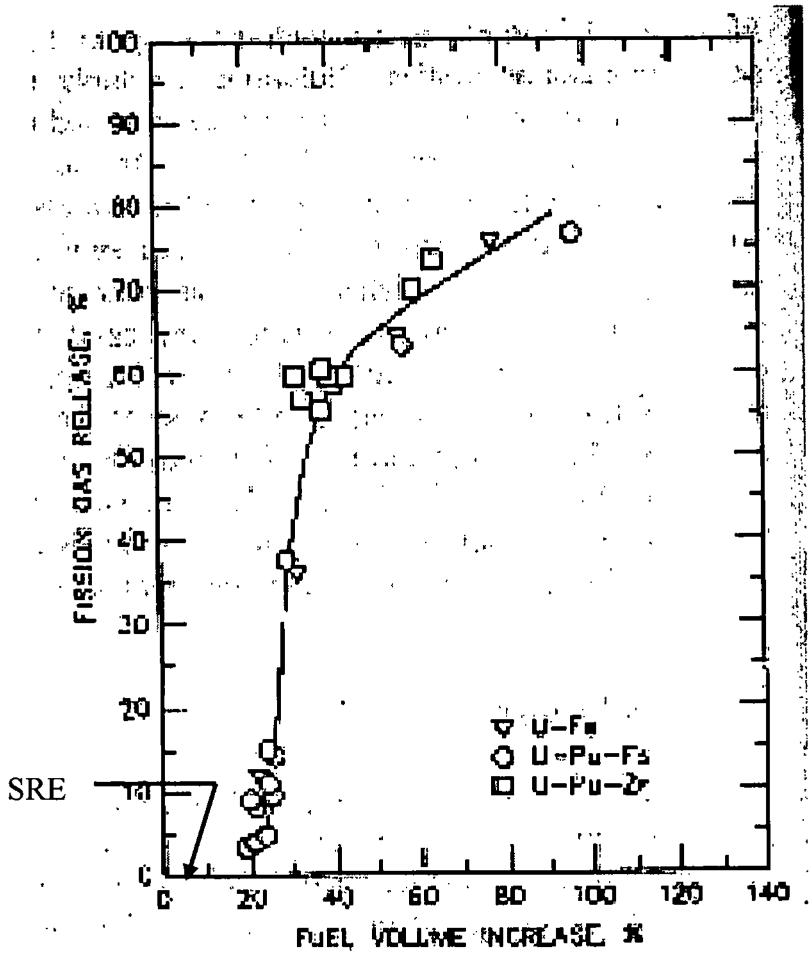


Fig. 3. Fission-Gas Release Versus Fuel-Volume Increase (Beck, 1968).

Figure 10. Fission gas release as function of fuel swelling in uranium metal alloy fuels[27, p. 90]

During the SRE fuel failure, additional swelling occurred as a result of the fuels cycling through the alpha-beta transition temperature of uranium, so portions of some of the fuel elements had higher swelling, but still substantially less than 10% on average. Measurements on three pieces of fuel rods showed some sections with increase in the maximum diameter dimension (they weren't round) to generally 0.850 in., including the cladding. The maximum increase noted was to 0.952 in.[6, p. IV-A-28] The original o.d. (outer diameter) was 0.79 in. This included a 0.010 in. gap and 0.010 in. thick cladding, so the uranium rod diameter was 0.75 in. Considering that the failed swollen fuels had no gap, subtracting the cladding results in swelling to 0.83 in. typical and 0.932 in. for the uranium, which correspond to typical total swelling of 10.7 percent in some segments and maximum swelling of 24%. As mentioned, this was in only one dimension and it didn't occur over the entire length.

While this still indicates, using the swelling relationship of Figure 10, that fission gases would not be substantially released from the fuel, the more important consideration is that this type of swelling does not represent pressure buildup from bubble formation that led to the relationship of Figure 10 and really would not have contributed to conditions of bubble release. The fission gases would not have coagulated into bubbles at the 0.1% burnup in SRE that would result in massive releases upon failure. These conservative estimates are below conditions for the occurrence of fission gas releases and explain why substantial fractions of the noble gases were not released to the cover gas in the SRE.

B. Experimental Breeder Reactor-II Uranium Metal Fuel

Chellow and Ader[34] studied the behavior of iodine during melt refining of irradiated synthetic EBR-II fuel. The fuel was uranium metal. The irradiated uranium was placed in a hole drilled in a fissium alloy and the charge was placed in a ceramic crucible and heated at a selected equilibrium temperature for several hours. The fissium alloy used had the average composition (weight percent): molybdenum, 2.4; ruthenium, 1.9; zirconium; 1.9; cerium, 0.6; rhodium, 0.3; palladium, 0.2; iodine, to 806 ppm (0.08 wt %); uranium, balance of 92.6%. The melting point of uranium is 2075°F (1135°C). The results of melt experiments showed that at 2282°F (1250°C) for 5 hours, less than 1% of the iodine was released. At 1300°C for 2.3 to 3 hours, 33 to 56 % was released, and at 1400°C at 3 hours, over 98% was released. No evidence was found that iodine vaporized as elemental iodine, I_2. The iodine acted like a metal iodide and was likely bound with uranium as triiodide, or with another fission product.[34] This interpretation is also provided by Monson et al.[35] This provides strong evidence that free elemental iodine (I_2) was not released from the SRE failed fuel elements that did not melt and never exceeded 1,093°C. In the Chellow and Ader experiments, the higher temperature (1,300 and 1,400°C) data were obtained in zirconia crucibles with impurities that were postulated to have reacted with uranium triiodide to form more volatile metal iodides. The 1,250°C experiment was done in a magnesia crucible where such reactions did not occur and the less volatile uranium triiodide remained stable.

X. DATA ON IODINE BEHAVIOR IN OTHER SODIUM-COOLED REACTOR METAL FUEL FAILURE OCCURRENCES SHOW THAT IODINE IS NOT RELEASED TO THE STACK

A. Fermi Reactor Metal Fuel Meltdown

The Enrico Fermi reactor had a sodium-cooled metal core like the SRE, of U-10wt% Mo, enriched to 25.6% in U-235. The fuel pins were clad in zirconium and were 0.158-inch outside diameter.[36, p. 13] On October 5, 1966, a broken off piece of zirconium baffle from the inlet area at the bottom of the core vessel resulted in flow restriction of the sodium and melting of one or more fuel elements.[37, pp. 31-37] This was a more severe condition than during the SRE July 1959 incident in terms of

- higher fuel temperature
- actual melting of the fuel, and
- severe boiling of the sodium around the failed fuel

all of which would have been more conducive to iodine releases from the fuel and through the sodium than in SRE, if Makhijani's theory is correct.

Qualitative and quantitative measurements of the fission products contained in the primary sodium coolant and the primary argon cover gas were made periodically after the October 5 incident. Where sufficient gas concentrations were available, analyses were performed with a 1,024 channel gamma ray spectrometer. When it was used, xenon-133 was identified as the major isotope present.[36, p. 80] Iodine-131 was not reported as having been observed. The major gamma peak energies and their probabilities of decay (abundances) for the xenon, krypton, and iodine isotopes of interest are summarized in Table IV.

Table IV. Gamma Spectra Peaks for Determining Isotopes in Gas Sample.[38]

Isotope	Maximum Peak Energy, MeV	Probability of Decay
Kr-85	0.513	0.00434
I-131	0.364	0.812
	0.080	0.0264
Xe-133	0.081	0.365

The I-131 peak falls between the Kr-85 and Xe-133 peaks in energy and its intensity (probability of decay) is greater. If it had been present in the cover gas, it would have been detected and quantified. Its presence was not determined. The only radioisotopes reported were xenon and krypton, both of which were used to estimate amount of fuel damage. Based on analysis of Kr-85 in the cover gas sample taken shortly after the operation on October 5, it was concluded that melting of the equivalent amount of fuel contained within about one complete subassembly was most likely, based on the assumption that all the krypton was released to the cover gas during melting.[36, pp. 80-91] The following evaluations of percent releases of fission products from the fuel are based on the assumption of the equivalent of one subassembly having melted. But, see next to last paragraph in this Section that updates this estimate to two equivalent subassemblies, so the following percents would be divided by two.

Analysis of the sodium showed the presence of cesium-137, strontium-89, iodine-131, barium-140, and lanthanum-140. The percents of fuel inventory of Cs-137 and I-131 found in the sodium were identical, similar to what was observed in SRE.[36, Table VII, p. 82] The conclusion was that about 1 or 2 percent of the available nonvolatile solid gamma-emitting fission products were released during melting. This is similar to the fractions of failed fuel inventories, including I-131, found in the SRE sodium. Also, the measurements indicated plate-out loss of approximately 93 percent for strontium-89 and strontium-90, 8 percent for cesium-137, and 75 percent to 84 percent for barium-140 and lanthanum-140 a few days after the accident and still increasing.[36, p. 81] This observation that all the fission products in the sodium were substantially plating out in the cooling system except cesium and iodine is the same as found in the SRE following its cladding failures. The cesium remains in the sodium because it is released from the metal fuel as elemental cesium metal or, possibly some as cesium iodide, CsI. When present as very low concentrations in excess sodium, thermodynamic calculations show that CsI will readily react with sodium to form NaI and elemental cesium. This is borne out by experiments by Castleman, Tang, and Mackay.[39, p. 382; 40] Sodium iodide is soluble in sodium and retained in solution at low concentrations.[39, p. 382; 41] Similar thermodynamic considerations show that uranium iodide in sodium converts to uranium and NaI. Cesium is very soluble in its sister alkali metal sodium.

In terms of the fractions of fuel inventories of cesium and iodine that were released from the molten Fermi fuel into the sodium, only a range can be estimated from data and statements give in the Fermi report. First of all, a radiation level of 2 mr/hr on the outside of the heat exchanger resulted from the plateout of gamma-emitting fission products. This was used to estimate a maximum of 1 or 2 percent of these solid gamma-emitting products was released during the melting, based on the assumption of the equivalent of one total subassembly having melted. This was inferred from the krypton-85 measured in the cover gas, assuming 100% release of krypton. Presumably, this 1 to 2 percent would apply to cesium. A subassembly contained 140 fuel pins. Table VII on p. 82 of the Fermi report lists the number of fuel pins corresponding to the quantities of Cs-137 and I-131 measured in the sodium. For 10% release, the Cs-137 and I-131 data corresponded to 162 and 153 fuel pins, respectively, virtually identical. Elsewhere, it was estimated that 8% of the cesium had plated out, so the numbers indicate 10.7 percent release of cesium, instead of 1 or 2 percent. Since I-131 and Cs-137 had close to the same values, one can conclude that between 1 and 10 percent of the I-131 in the melted pins was released to the sodium. Given that the fuel melted, as much as 10 percent is not unreasonable. It more than brackets what would have been released to the sodium in the SRE incident, where about 1 percent has been deduced. And, as noted above, none of the iodine in the Fermi reactor (nor SRE) got to the cover gas.

Similar Fermi data assessment indicates that about 15% of strontium-90 was released to the sodium.

An apparent later evaluation of the data concluded that the best estimate of amount of fuel melted was the equivalent of two subassemblies rather than the one assumed in the above reported analyses.[42] Thus, the percent releases are apparently one-half those originally derived and presented above. Then, the percent iodine release from the fuel would correspond to 1 to 5 percent.

These observations from the Fermi fuel melting incident are consistent with observations of the SRE incident that show that no I-131 reached the cover gas and, just as significantly, the amount of I-131 captured by the sodium was similar to cesium, only 1 to 5 percent. The fact that only a fairly small fraction of iodine was found in the sodium and none in the cover gas demonstrates that little iodine was released from the melted uranium. The conditions here would have been more conducive to iodine release from the fuel and, also, to bubbling through sodium into the cover gas than in the SRE that is suggested by Dr. Makhijani. Iodine-131 was not found in the Fermi reactor cover gas nor substantially in the sodium.

B. EBR II Fuel Failure Incident

The Experimental Breeder Reactor II at the Argonne National Laboratory site in Idaho Falls, (EBR II) was a sodium-cooled reactor with a sodium bond between the metal fuel and cladding of the fuel pins, similar to the SRE (which had a similar alkali metal bond of sodium-potassium, NaK). On May 24, 1967, an experimental fuel experienced failure. The reactor was equipped with two types of detection systems to monitor release of fission products from a failure. One, called FERD (Fuel-Element Rupture Detector) monitored delayed-neutron-emitting fission products, of which iodine is one, in the sodium. The other, called CWD (Charged Wire Detector), monitored the short-lived rare gas (xenon and krypton) fission product daughters in the cover gas on a continuous basis.[43,50] The combination of measurements with the two instruments determined which of two failure modes are recognized as being possible with sodium-bonded fuels, depending on the location of the defect with respect to the fuel. For a gas-type failure, one occurring in the gas region above the fuel, rare gas fission products will be released directly to the primary coolant and will diffuse upward into the cover-gas plenum. For such failures, iodine and bromine are not released, but remain chemically fixed in the sodium bond, which remains intact.[43]

For a bond-type failure, one occurring in the vicinity of the fuel, bond sodium containing delayed-neutron-emitting species (including iodine) along with gaseous fission products will be released directly to the coolant. Such failure will be annunciated by both monitoring systems.

For the fuel failure incident on May 24, only the CRD indicated fission noble gas releases. The FERD did not detect delayed-neutron emitters, including iodine, in the sodium. This demonstrates that, indeed, the iodine was not released as elemental gas (I_2) from the top of the fuel plenum along with the noble gases into the sodium. The iodine either remained in the fuel or it was reacted with and retained by the sodium bond.

This provides evidence that, if elemental I_2 is released from fuels, the alkali metal bond will react with and retain it as sodium iodide and potassium iodide. If a failure releases the bond material into the sodium, the iodide would remain in the sodium.

C. EBR-II Run-Beyond-Cladding-Breach Tests

Complementary results to the plenum failure incident that further elucidates the release behavior of fission products from metal fuels were obtained from several so-called run-beyond-cladding-breach (RBCB) tests performed in EBR-II.[27, pp. 105-106; 44] In the tests, fuels were pre-irradiated to high burnup. Then, the cladding was thinned and the fuels reinserted into the reactor, shortly after which the cladding failed. Irradiation continued for 54 to 233 days after cladding failure for the different tests; final burnups were very high, ranging from 6.0 to 14.4 percent. By comparison, SRE fuel burnup was at 0.1%. The responses of the FERD and cover gas instruments were recorded. The FERD gave a short response immediately after failure. This release corresponded to the bond sodium release. The cover gas instrument, which measured the noble fission gases (but not I-131) in the cover gas, gave a similar release signal concurrent with the FERD. Recall that the FERD detects iodine along with other fission products in the sodium. A large portion of Cs-133, the decay product of Xe-133, was expelled with the bond sodium.

Thus, previously released Xe-133 into the gap decayed there and the cesium was dissolved into the sodium bond. After the bond sodium was gone, transport of delayed-neutron precursors, which are detected by the FERD, to the breach site was slowed down enough that they decayed before reaching the reactor coolant. This is an important observation. The FERD does not distinguish the specific type of delayed neutron fission products, so one can't say whether or not I-131 was present with other fission products in the released bond sodium. But, the fact that I-131 was not released subsequently to the bond sodium release shows that, if it were in the fuel gap prior to cladding failure, it would have reacted with the bond sodium and was not released as I_2 into the coolant upon rupture and was not released before decaying after rupture. And, these are at extremely high burnups where gases would be released. Thus, iodine apparently was not present in and released from the fuel as gaseous I_2. This is pertinent to the situation in SRE – it is another experiment that proves elemental I_2 would not be released from the very low burnup SRE metal fuel.

According to an author of the report, additional release of noble gases following the rupture release occurred only to the extent of recoil from current fission production.[32] Recoil ejection occurs for fission products born within a few microns (micrometers) of the surface.[45] The fraction depends on the surface-to-volume ratio. For SRE fuels, the theoretical release fraction is about 0.035%.[6, p. IV-C-6] The significance of this will be seen later when evaluating Dr. Makhijani's theory for the SRE reactor.

This, again, demonstrates that iodine is not released from the metal fuel as it is irradiated, even at very high burnups that result in noble gas bubble formation in and release from the fuel. This is further experimental verification that iodine exists as non-volatile salts in the metal fuels.

D. International Experience and Safety Designs of Liquid Metal Fast Breeder Reactors Recognize and Credit Retention of Iodine by Sodium

In 1973, an expert report on control of iodine in the nuclear industry, in reviewing state of knowledge and experience to provide recommendations for iodine controls in a sodium-cooled reactor, stated that controls in the off-gas system for radioactive iodine-131 were not needed. "Thus, it would be expected that essentially all of the iodine entering the primary coolant would immediately react with the sodium and would be retained within the primary system. Experience with operating LMFBRs indicates that is in fact the case....Because of the retention of iodine by sodium, special equipment for removal or control of iodine is not required for an LMFBR off-gas system. A high-efficiency particulate filter system for the removal of sodium aerosol provides adequate control of the radioiodine, even in the event of substantial cladding failure.[46]

This is in reference to oxide-fueled reactors where fuel failures involving melting at extremely high temperatures would release elemental iodine, I_2, a worst case not applicable to the SRE metal-fueled reactor. Safety designs are always extremely conservative and this recommendation considered all known data at the time, both laboratory studies and experience of operating reactors.

XI. IODINE RELEASED FROM FUEL CAN REACT WITH AND BE RETAINED BY STAINLESS STEEL CLADDING OR BY NaK BOND

An additional possible mitigating reaction of iodine with stainless steel cladding was experimentally characterized by Aubert et al.[47] Under isothermal conditions, iodine reacts directly with 316 stainless steel forming the iodides MnI_2, CrI_2, FeI_2, NiI_2, or MoI_2. In a thermal gradient, "spectacular corrosion of the steel at low temperature is accompanied by substantial loss of chromium." The corrosive action of iodine on steel does not require the presence of oxygen. "In irradiated oxide fuel, free iodine (or iodine liberated by the decomposition of cesium iodide, a compound of fission products) can take part in the development of the reaction between fuel and cladding."

The extent of this reaction in quantitatively taking up iodine in either the elemental or metal iodide form cannot be predicted from this information. The thrust of the research was to determine the effect of iodine on the corrosion of steel. The results indicate that if iodine were to be released from the uranium metal fuel (which other data and theory show did not happen in the SRE July 1959 incident), a substantial quantity could be trapped by the cladding.

The assistant editor of Nucleonics journal summarized the symposium on "Fission Product Release and Transport under Accident Conditions," held at Oak Ridge, Tennessee April 1965.[48] In citing a paper by Collins[49], he states "When iodine is released from stainless steel or Zircaloy-clad UO_2 at high temperature, when in carbon dioxide or in steam, the largest fraction attaches itself to solid particles derived from the molten cladding material." Here, the situation is relevant to the SRE cladding failures because some of the cladding melted by alloying with uranium. The iodine from the UO_2 fuel in the above study would likely have been largely in elemental form that Makhijani claims, though incorrectly, for the SRE fuel. This study does indicate that, if I_2 were released from the metal fuel, it would have substantially reacted with the molten cladding as it alloyed with the uranium in the eutectic formation.

Finally, in order for substantial fractions of iodine to be released from the fuel matrix as elemental I_2 gas to the fuel gap and be available for rapid release upon cladding failure as Makhijani postulates, the postulated I_2 gas would have had to diffuse out in advance of the cladding failure and been in contact with the NaK (sodium-potassium alloy) liquid bonding material in the gap for considerable time. Since I_2 reacts almost instantaneously with hot, liquid alkali metals such as Na (sodium) or NaK, (and plaintiff expert witness Makhijani acknowledges that iodine (I_2) has a high solubility in and reacts rapidly with sodium[65, p. 78) it would have quantitatively converted to sodium iodide (NaI) and potassium iodide (KI) while in the gap.

This is recognized by others, as well. In describing the FERD in EBR-II for detecting fission products in the sodium coolant resulting from failed cladding (see Section X.B), Smith et al.[50. p. 19] state, "Underlying this conclusion is the knowledge that relatively long-lived species such as I-133 and I-135 accumulate in the sodium bond of normal driver elements and in the sodium bond between the damaged cladding of an experimental fuel element and its capsule." These so-called "relatively long-lived" isotopes have half lives of 20.8 hours and 6.57 hours, which are quite short relative to the 8 day half life of I-131. This indicates rapid reaction of iodine with the bond material.

The fact that the quantity of iodine found in the sodium coolant of SRE that would have included the NaK and its constituents was only a few tenths percent of the failed fuel inventories indicates, simply, that the iodine did not substantially release from the metal fuel.

XII. EXPERIMENTS OF IODINE BUBBLES IN SODIUM SHOW THAT ELEMENTAL IODINE REACTS WITH AND IS NOT RELEASED FROM SODIUM

Hart performed experiments to determine the degree of retention in the sodium and/or movement to the reactor cover gas system of several fission products.[51, 52] The nuclides Cs-137, I-131, and Sr-89 were selected for study in part because they could have high volatility at high temperatures. He added iodine in the form of sodium iodide salt to a columnar tube containing sodium. Clearly, this is because he believed (correctly) that any elemental iodine (I_2) escaping from fuel would be trapped in the sodium by reaction to form sodium iodide. The sodium was heated to 1525°F (829°C), with successive sodium and gas samples taken at 100°F intervals above 900°F. Three tests were conducted and in every case less than 0.001% of the iodine radioactivity was found in the gas phase. The importance of this test is that it showed that iodine, if present in the sodium, will not escape and be released to the cover gas.

A more significant experiment was one in which bubbles of vaporized elemental iodine, I_2, were released into sodium at elevated temperatures.[53, pp. 22-26] Crystals of iodine (I_2) were encapsulated with or without diluent nitrogen gas. The capsules were immersed in a twelve-foot high by four-inch diameter column of sodium at depths of 6 feet or 10 feet below the surface of the sodium. After heating to 500°F (260°C) the capsule was punctured and the iodine released to the surface was swept through a charcoal filter for capture and analysis. The experiment with no nitrogen and six-foot immersion released only 0.2% of the iodine from the sodium. Six feet is the depth of sodium above the top of the fuel elements in the SRE reactor; the six-foot long fuel elements extended further down in the sodium. This is a conservative experimental design because the iodine had no opportunity to disperse laterally as it could in the reactor sodium pool and experience longer residence times, which would give the iodine more time to react with sodium. When nitrogen was present to cause a gas bubble, the iodine release from six feet was 1.0 and 1.5% in two tests and from ten feet (more representative for the submerged fuels) it was 0.2%.

Tests were conducted at 1000°F (538°C), as well. At this temperature, sodium vapor carryover to the charcoal filters increased the minimum detection limit for I-131 and it was not detected in any of those tests (detection limit ranged from 0.002% to 1.4%). The 1000°F tests were confounded by the fact that air leakage into the cover gas region formed an oxide layer on the sodium surface that may have tended to inhibit the release of iodine bubbles to the cover gas region.

The reaction rate of gaseous iodine (I_2) with hot liquid sodium increases exponentially with temperature. A rigorous relation is that reaction rates increase exponentially with temperature according to the Arrhenius rate law:

$$k=Ae^{-E^*/RT}$$

The reaction rate constant, k, is temperature dependent. A is a constant that cancels when comparing two temperatures. R is the universal gas constant, 1.9872 calories deg^{-1} mol^{-1} when E^* is expressed in calories mol^{-1}. The temperature effect is expressed in the value of E^*, called the activation energy. That is the energy barrier that the colliding molecules must overcome before combining. Since the reaction rate of iodine with liquid sodium is so fast, its activation energy hasn't been characterized. Typical activation energies, E^*, range from 25,000 to 45,000 cal/mol. An example for a gas-metal reaction is 46,000 cal/mol for nickel reacting with molecular oxygen and 27,000 cal/mol for nickel reacting with atomic oxygen.[54] If one take the mid point of the activation energies as a probable value for providing a temperature effect (although the molecular oxygen, O_2, case would be more representative for I_2 – and result in much greater temperature effects), one calculates the reaction rate at the boiling point of sodium, 1,621°F, would be more than one hundred million times faster than at 500°F, which is very fast to start with.

Thus, at near the boiling temperature of sodium when some of the fuel claddings failed, the kinetics of the reaction, relative to that at the 500°F tests here would be immeasurably fast. The rate would be limited only by the rate of mass transport of iodine in the bubble to the bubble/sodium interface. Again, at the boiling temperature of sodium, both diffusion and convection forces would make this extremely rapid and, in particular, much more rapid than at 500°F.

Equally important, the bubbles, if formed, would be saturated with sodium vapor molecules at the temperature of the sodium. [See Bubble Hydrodynamics web paper: http://www.bubbleology.com/Hydrodynamics.html. In describing bubbles in water, the author states "The water molecules at the interface are continually exchanging with water molecules in the gas (according to the equilibrium vapor pressure)."] The vapor pressure of sodium at 500°F is 4.34×10^{-6} atmosphere and, at the boiling point in SRE, 1,621°F, by definition, 1 atmosphere. The corresponding vapor concentrations are 6.0×10^{13} molecules/cc at 500°F and 6.3×10^{18} molecules/cc 1,621°F. The sodium vapor concentration in boiling sodium is 100,000 times that in the 500°F experiment.

Plaintiffs' expert witness Makhijani suggests[65, p. 80] that I_2 released from the fuel is simultaneously diluted by xenon and krypton to less than 1 mole percent as it is inserted into the SRE boiling sodium. Though Dr. Makhijani does not recognize and discuss it, the sodium vapor atoms in the boiling sodium bubbles would further add to the total gas molecules, much more than the noble gases. Therefore, the sodium atom concentrations would be in great chemical excess relative to iodine. But, in the 500°F experiment, the sodium atom concentration in the bubbles would be hundreds of thousands times *less* than the iodine concentration. In addition, as we just saw, the reaction rate constant at boiling would be many more than 100 million times greater at the boiling temperature than at 500°F. The sodium vapor atoms would be rapidly reacting with the iodine molecules in the bubble as it traverses to the top. This effect would be virtually infinitely greater for the SRE bubbles at 1,621°F than for the experimental bubbles at 500°F, both because of the much greater sodium atom concentration and the much greater reaction rate constant at the higher temperature.

The boiling sodium just as it exited the assemblies would be at about 1,686°F where the kinetic reaction rate effect was evaluated above as greater than 100 million times relative to the 500°F iodine-sodium capsule bubble experiment. As the sodium moved toward the top of the pool, the temperature would decrease and at the top, the surface temperature was about 542°F. The maximum exit temperature measured from a fuel channel during the reactor excursion on July 13 was 755°F, which was for the Channel 10 failed element.[6, p. II-12] Thus, while the overall kinetic effects would be decreased from those at the boiling temperature, they would have still been overwhelming compared to the static 500°F temperature of the sodium release experiments, still starting at 1,686°F for a brief period that would have been sufficient for reaction to complete, but at the exit temperature of 755°F, rates would have been more than 1,000 times faster than at 500°F. At the surface 542°F temperature, it would still have been more than four times faster.

It is clear, then, that the 500°F tests bracketed the SRE conditions. These tests demonstrate that had elemental iodine vapor (I_2) been released from the failed fuels into the sodium as theorized by Makhijani, it would not have escaped to the cover gas. They are consistent with the interpretation regarding the absence of observed iodine in the cover gas: "The incident of the fuel damage to the SRE also provided data on sodium chemistry. Analysis of the cover gas revealed no fission products other than the noble gases. A search for I^{131} failed to reveal its presence in the cover gas; this indicates, as was expected, that iodine and bromine fission products immediately combine chemically with sodium."[4, p. 78]

Another capsule rupture experiment was even more vigorous and showed less than 0.7% of I-131 released into sodium was released to the helium cover gas.[55, pp. 34-37; 56] The experiments were designed to provide a more realistic insertion of fission product activity into sodium to determine the retention characteristics of a bubble of iodine and fission gas being transported through a sodium column. A 25 cm^3 capsule containing a thin foil (1.5 mil, or 0.038 mm thick.) of uranium in an atmosphere of helium was irradiated. It was fitted with a rupture disc and inlet connected to a "firing" chamber of argon gas. The irradiated capsule was immersed to a depth of 6.3 inches in 5.5 liters sodium at 1000°F. The firing chamber admitted argon gas and the rupture disc released the gases at 100 psig. When the pressure was relieved to about 1 atm, the volume of the bubble would be 150 cm^3 or more (depending on volume from the firing chamber). About 15% of the calculated I-131 inventory was released into the sodium and 0.1% in the cover gas, which corresponds to 0.66% of that in the sodium. This, again, is proof that a large bubble of noble gas with iodine does not prevent almost complete reaction or retention of the iodine in sodium, even in this small depth. This was an extremely vigorous condition. The bubble would have been thrust with great energy into the sodium. The experiment certainly brackets Makijani's bubble scenario in SRE.

A related experiment in which irradiated uranium metal fuel was melted under sodium is relevant.[57] The fission products released from the melted fuel that was maintained molten for three minutes were released into a cylinder approximately three feet tall containing 120 L of sodium at 1,022°F (550°C). Sample points in the sodium column were provided at six different levels. When the sodium was stagnant, the I-131 collected at the bottom of the sodium column. The I-131 concentration in sodium fell off with height according to

$$A_S = A_{S0}\, e^{-3h}$$

where A_{S0} represents the specific activity in μCi/g at zero level and h the sodium height in meters. Thus, at two meters (a little over six feet, as in the SRE above the core), the fraction of iodine was $A_S/A_{S0} = 2\times10^{-8}$, which shows that the I_2 reacted and was removed as sodium iodide, NaI, before it reached that point.

When argon gas was injected into the sodium at the bottom near the point of the melted fuel, the turbulence resulted in uniform concentration of I-131 throughout the sodium.

The argon cover gas was passed through a "May pack" filter that contained activated charcoal to trap any iodine. None was ever found in it. About 0.001 to 0.01% of the iodine was found condensed with sodium and it was transported in the form of sodium iodide.

These experiments demonstrate that elemental iodine, I_2, is not released from uranium metal fuel and transported through sodium into the cover gas, even with violent bubbling and turbulence created with argon injection, a condition similar to Makhijani's "bubble theory".

In the SRE the helium supply from the pressurized gas cylinders was purified by bubbling the gas through NaK (sodium-potassium liquid at room temperature) to remove oxygen to low levels before it was routed to the gas services.[14, pp. 175 and 179; 58] This author has used a NaK bubbler to similarly purify a helium stream to extremely low levels of oxygen in laboratory experiments. These provide further evidence that trace levels of impurity gases in bubbles in the liquid alkali metal readily react with the liquid metal as the bubbles pass through it. In these cases, the reaction occurs at ambient temperature where all kinetic effects (rates, both chemical and mass transfer) are much, much less than at the boiling point of sodium.

XIII. WHEN DID THE FUEL CLADDING FAILURES OCCUR?

A number of different indicators provide general information about when the fuel cladding failures may have occurred. While the exact days in Run 14 that cladding failures and fission product releases cannot be completely pinned down, the predominant evidence is that most of the failures occurred on or about July23, with a small fraction of the failed elements having been damaged about July 12 and July 13. An overview is provided here of some different perspectives.

Activity was observed in the high bay on July 12, but corresponds to a very tiny amount of curies. Stack activity on that day did not coincide in time with the release from the cover gas by pressure venting, indicating that the cover gas was directed to the decay tanks. The stack activity has been shown by Daniel to have originated from operations of the fuel handling unit, which released only one cubic foot of gas at a time.[59] Clearly an element or two may have failed then, but not all thirteen.

Based on activity in the gas storage tanks continually decaying following July 15, it was concluded by the AI investigators that most of the fuel damage must have occurred just after the start of Run 14, probably within the first three days. [6, p. IV-C-16]. However, this statement that the activity continually decayed after July 15 is not supported by Borg's reported activities in the gas decay tanks during releases.[60] It shows the following activity concentrations on succeeding dates:

Sample Date	Activity, $\mu Ci/cm^3$
7/11/59	2.2×10^{-6} (normal)
7/20/59	5.0×10^{-2}
7/25/59	1.4×10^{-2}
7/25/59	8.0×10^{-2}
8/22/59	7.0×10^{-2}

A more complete compilation and correlation of activity measurements in the cover gas and decay tanks with potential fuel damage dates and cover gas venting operations is summarized in Table V.

It is clear that some fuel damage with noble gas releases occurred in the July 12-13 time frame and, also, after July 20 – the latter date because the August 1 cover gas sample compared with the August 12 sample indicates the presence on August 1 of short-lived isotopes., of which Xe-133 would be the longest lived. Those two data points, August 1 and August 2, are inconsistent with a July 13 release from fuels but are consistent with a July 23 release. These data taken in combination with other evaluations below point to conclusion that the majority of fission gas releases from fuels occurred at the later date. The decay tank release activities would have occurred following cover gas venting by some time after shorter-lived isotopes, including Xe-133, had decayed to permissible levels for discharge.

A comparison of sodium and cover gas analytical data for fission products with the fission product inventories of the thirteen failed fuel elements can provide some indication of when those fission products in the sodium were formed in the fuels. Appendix A describes the derivation of the percent releases described in the following. The first sodium sample was taken on August 2, seven days after shutdown and three weeks after a suggested release date of July 13.[1, p. 11; 6, pp. IV-C-2 to IV-C-4] A second sample from October 31 showed that all fission products in the sodium except cesium and iodine substantially plated out over time.[1, p. 12] This may be understood by the facts that the cesium (mostly elemental, see Section VI), being very similar to its alkali metal partner, sodium, is highly soluble in sodium, as is iodine as sodium iodide [41] that forms by reaction with sodium, regardless of its originating chemical species.[*] Thus, the comparison of those two fission products may provide some indication of the release date inventory. If one assumes a July 13 release, the sodium data indicates 1.1% release of cesium-137 and 1.3 % release of iodine-131. If one compares the sodium data with July 23

---

[*] Thermodynamic modeling indicates that all potential forms of iodine in the metal fuel – UI_3, CsI, other fission product iodides – at the quantities present would all convert to sodium iodide, NaI, in the sodium pool. The same is of course true of elemental I_2, although I do not consider it a potential form in the fuel. The total iodide from the thirteen failed elements amounts to about 0.013 g-atoms (see Table II). About 0.3 to 1% (depending on the assumed date of failure) of the inventory was measured in the sodium, which would dissolve to a concentration of 6×10^{-9} molar in the 27,600 L of sodium in the core plus piping and heat exchanger, well below the solubility at 344°F (260°C), the normal inlet temperature to the core.

Table V. Time sequence of operations and activity concentrations in cover gas and decay tank samples.

Date	Operation	Cover Gas Activity[*]	Decay Tank Activity[*]
July 11			2.2×10^{-6} μCi/cm^3 normal
July 12	Fuel failure. Vented 7.7%		
July 13	Fuel failure?		
July 15	Vented 19.6%		Hot after 2-3 days, several mR/h; >>1 μCi/cm^3
July 18		Hot after 5-7 days, 30 mR/h, 5 days after suggested fuel failure[**]	
July 19 & 20	Vented 19.6%? (Pressure tests on reactivity)		
July 20			0.05 μCi/cm^3
~July 23	Fuel failures		
July 25			0.014 μCi/cm^3 0.080 μCi/cm^3
July 29	Vented 18%		
Aug 1		5.5 μCi/cm^3, 8 days after postulated fuel failure	
Aug 12		0.87 μCi/cm^3 (~0.79 μCi/cm^3 Xe-133 = 2.0 μCi/cm^3 on **Aug 1.**) $\therefore$, **short-lived isotopes then.**	
Aug 14		0.87 μCi/cm^3	
Aug 22			0.070 μCi/cm^3

[*] Activity concentrations are expressed for volume at standard temperature and pressure, STP, i.e. 1 atmosphere, or 14.7 psia.

[**] Short-lived isotopes with half-lives of a few hours to days were the cause of the moderately high radiation reading.

calculated fission inventories in the fuels, the calculated releases are 1.1% of cesium-137 and 0.3% of iodine-131. This might tend to support the conclusion of early releases, if one assumes similar release fractions for the two. This assumes that the iodine and cesium were released by the same mechanism into the sodium, viz., leached from fragment portions of the uranium fuel. While they may tend to favor the early release model, these data have enough uncertainties in assumptions and mechanisms that they cannot be taken alone to prove the assertion. Cesium would have been exposed to the sodium as elemental cesium while iodine would have been in the form of uranium iodide, UI_3. Differences in dissolution rates of the two different structural forms could account for the differences based on a July 23 release. The UI_3 molecular concentration in the uranium matrix would have been at 1/100th the concentration of cesium and this could affect the dissolution, making the UI_3 more difficult to get into solution (dissolve slower). Thus, the July 23 relative percents of fuel inventory for iodine and cesium in sodium is not unreasonable and would not rule out a July 23 majority release.

The analytical results for Xe-133 and Kr-85 in the cover gas sample taken on August 12 and analyzed on September 14, when extrapolated back for decay from possible operating release dates can be analyzed in a similar manner. The calculated release percentages indicate that, based on the July 13 inventories calculated by Daniel & Associates[8], the percent Xe-133 release (about 4.3%) would be 70 times the percent Kr-85 release (0.062%), a seemingly unreasonable result for the two gases that had very similar atom concentrations in the uranium (see Table II). Based on the July 23 inventories, the percent releases would be about 0.18% of Xe-133 and 0.038% Kr-85, a ratio of 4.7. This favors releases towards the end of Run 14. The differences of this magnitude between Xe-133 and Kr-85 may be explained largely by the difficulty of measuring small activities of Kr-85 in the presence Xe-133. Typically, errors of this magnitude are experienced for Kr-85 and always on the low side.[61] The overarching conclusion is that the apparent result of about 70 times the Xe-133 percent release relative to percent Kr-85 release based on an assumed July 12-15 release is extremely strong evidence against that release date.

The summary of these results, along with those for Cs-137 and I-131 in the sodium, are displayed in Table VI, taken from Appendix A.

Table VI. Calculated percent releases of fission products from failed fuels for postulated release dates of July 13 and July 23.

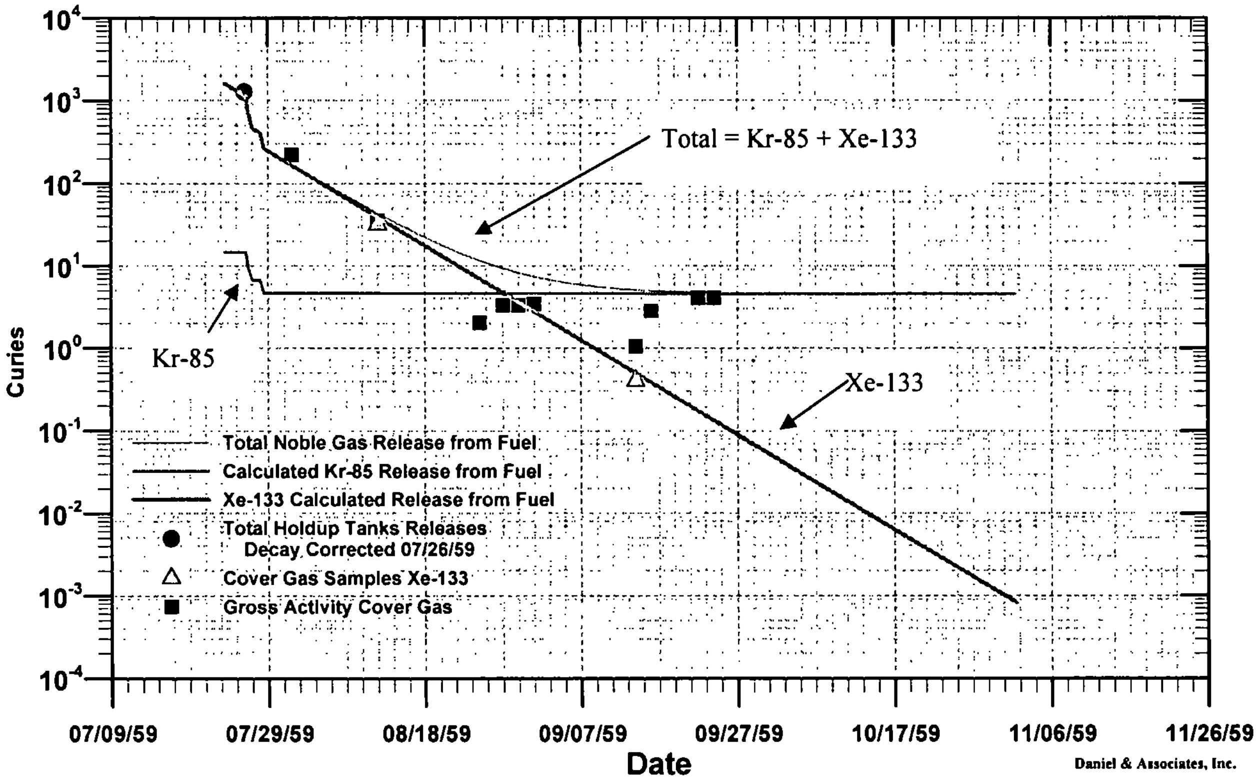
Isotope	Calculated Percent Released from Failed Fuels	
	July 13 Release Date	July 23 Release Date
Xe-133	4.3	0.18
Kr-85	0.062	0.038
I-131	1.3	0.28
Cs-137	1.1	1.1

An error in Kr-85 analysis is also indicated by comparing these gamma spectroscopy results for the August 12 cover gas sample with the independently measured gross activity of the sample on that date. The gross activity measurement was 0.87 $\mu Ci/cm^3$.[6, p. IV-C-14] When the concentrations of Kr-85 and Xe-133 measured in the sample on September 14 by gamma spectroscopy are extrapolated back to August 12, the result is 0.78 $\mu Ci/cm^3$ Xe-133 and 0.016 $\mu Ci/cm^3$ Kr-85, a total of 0.80 $\mu Ci/cm^3$, instead of 0.87 $\mu Ci/cm^3$. The two independent values can be made to match if the Kr-85 concentration is multiplied by 4.6. This is virtually the same as the 4.7 ratio of percent release Xe-133 relative to percent Kr-85 release calculated for a July 23 release based on the gamma spectroscopy measurements and inconsistent with the ratio of 70 for a July 13 assumed release date. By multiplying the Kr-85 concentration by 4.7, the Xe-133 and Kr-85 percent releases calculated for a July 23 release are identical and it make the sample results consistent with the gross activity measurement.

It is not particularly surprising that the calculated noble gas percent releases are less than those of other fission products. There are a couple of possible contributing reasons for this. One is different mechanisms of release from the fuel matrix. The non-volatiles (including iodine as a metal iodide salt) would have dissolved into the sodium by virtue of the sodium interacting with the fuel. The noble gases, on the other hand, do not interact chemically with sodium (their solubilities are extremely low[62, 63, 64]). As described in Section IX.A, at the very low burnup of the SRE fuels of 0.1%, the gases have not agglomerated into bubbles that can be readily released from the fuel matrix.

Another potential reason may be that the cover gas sample was not entirely representative of the core cover gas region. Hart indicated that the sample was representative in his statement, "Direct samples from the primary pool blanket gas may be taken by drawing the helium into an evacuated container from a suitable connection, usually the helium fill line."[1, p. 5] Furthermore, he reports the sample results in $\mu Ci/cm^3$ along with the calculated total curies in the cover gas, from which one calculates a cover gas volume of 6,300 L, which would correspond to the core cover region and would not include the 33,300 L in the connected sodium fill tank. The core cover region is connected to the fill tank by 50 foot run of 2-inch diameter pipe, which would pretty much isolate the gases in the two. The helium fill inlet lines are between the reactor core cover region and the sodium fill tank. Valves between the two vessels are open during operation. But, the samples were taken after shutdown and the valves may have been closed, isolating the fill tank from the core region. However, gas in the connecting line between the sampling point and the core region could possibly have diluted the concentration of the core gas contents some as they were drawn into the 2 liter evacuated sample bottle. The interim report notes, "The cover gas is sampled at a location downstream from the core (rather than at the reactor loading face) using an evacuated (~2,000 cm^3) chamber."[6, p. IV-C-13] How many drawings were done to get a representative sample is not known. If the sample were more representative of the average of the entire cover gas system, including the fill tank, then the measured concentration would be multiplied by about 39,600 L instead of 6,300 L and result in about a factor of 6.3 greater quantities than tabulated for Xe-133 and Kr-85 in Table VI. This is likely an outside error and would place the Xe-133 and Kr-85 release percents in Table VI for a July 23 release from fuel at 1.1 and 0.24 percent, respectively, quite in line with the sodium fission product data for I-131 and Cs-137. The probable value likely lies between the two extremes. As described in the next paragraph, comparison of the holdup tank data with the cover gas data indicates that multiplying the cover gas activity concentrations by the total cover gas system volume, including fill tank, results in closer alignment with the holdup tank data. Any absolute errors in the quantity would not affect the ratio of Xe-133 and Kr-85 activities in the cover gas sample that are important in the evaluations of release date.

A relevant and important evaluation of cover gas and decay tank gross activity measurements as a function of time points strongly to a later release. This evaluation is in the report of J. Daniel.[59] His results are significant in showing that a July 23 release date is consistent with bringing independent gas measurement data together and are reproduced here as his Figure 5.5 below. The solid lines represent the number of curies of Kr-85 and Xe-133 and associated short lived isotopes that would have been in the cover gas based on the measured gross activity amount released from the decay tanks and the ratio of the Kr-85 and Xe-133 isotopes in the failed fuels on the date of the assumed release, July 23. The solid curves as a function of time represent the radioactive decay as well as estimated pressure ventings of the cover gas so that the two different sources of data can be compared to each other. The black squares represent the completely independent measurement of gross activity concentration in the cover gas[6, Table IV-C-8] multiplied by the cover gas volume. Daniel used the total cover gas volume of core region plus fill tank in converting concentrations in cover gas samples to quantities. If the inventories of Kr-85 and Xe-133 in the fuels are the correct amounts (and they vary with dates) the two different sets of data for total activity should align together on the graph. In particular, the curve for the total Kr-85 and Xe-133 must intersect the total holdup tank releases decay corrected to July 26 and then follow the decay measured in the cover gas samples from that point on. One sees that is the case for the July 23 release. Effects of flushings beyond August 26 are not included because of difficulty in quantifying them. If known and applied, it would lower the lines at those later dates a bit and bring them more in align with the cover gas data points in that region for the July 23 release.



Daniel Figure 5.5. Cover gas total activity decay curve – July 23 release date.

This is quite strong evidence that the majority of releases occurred about July 23 and not July 13 – especially when considered in combination with the cover gas gamma spectra results presented above that show wide discrepancies between krypton and xenon based on an assumed July 13 release and with the fuel temperature data (see below and Section VII) that show cycling through the α-ß phase of uranium at the later date.

Other data that are relevant are the dates of fuel and sodium temperature excursions and cycles. This is discussed in Section VII. The evidence there is that temperature conditions, and cycling of temperature through the alpha-beta temperature that would cause fuel swelling were prevalent from July 22-24 and less so July 12-15. And, the temperature of R-55 from July 23 to shutdown on July 26 (three days) did not decrease back to the level of the July 15 temperature (two to three days of any postulated temperature excursions). This indicates an unlikely temperature excursion between July 12 and 15.

Direct evidence of fuel failure after July 22 was obtained when some fuel elements (assemblies) were "jiggled" on July 25 in an attempt to dislodge some of the material believed to be interfering with coolant flow. It was discovered then that fuel elements in core channels 10, 12, 35, and 76 were stuck.[6, p. III-17] But, on the evening of July 22, a similar operation showed that the element in core channel 10 was free. Thus, it failed between those two dates. It should also be noted that the moderator can in core channel 10 is one that Dr. Makhijani claims provided a pathway for iodine release from that assembly on about July 13 without traversing through the sodium pool. These observations prove that such an unencumbered release from element 10 did not occur.

All this evidence taken together indicates definitively that most fuel failures occurred toward the end of Run 14 and I accept this as the prevailing situation.

The worst case, in terms of assessing effects of releases of I-131 vis-à-vis Dr. Makhijani's multiple hypotheses, which rely on release to the stack on July 15 during a venting operation, would be majority fuel failure and fission product release during the first three days of the run, before July 15. In order to show that the operating data prove that Dr. Makhijani's theoretical model is incorrect, most of the analysis here of the data is performed assuming a majority of release during the July 13 excursion and a small amount on July 12. Arbitrarily, 10% on July 12 and 90% on July 13 is assumed, although the exact split is quite inconsequential. The subsequent analyses of data on this basis will show that, in addition to the chemical reasons and barriers against I_2 release, even if these were the primary release dates as suggested and relied on by Dr. Makhijani, very little of his theoretical elemental I_2 gas would have been released to the stack. One should not infer that, by modeling on the basis of majority releases from the fuels before July 15 as suggested by Makhijani, I accept that to be true. Quite the contrary, I believe the weight of evidence is for later releases.

XIV. PRESSURE CHANGES AND VENTING OF COVER GAS DID NOT QUANTITATIVELY "FLUSH" GASES OUT PRIOR TO SAMPLING – IODINE-131 WOULD HAVE BEEN MEASURED IN THE AUGUST 12 SAMPLE WERE IT INITIALLY PRESENT

Plaintiff expert witness Makhijani asserts that pressure venting operations would have removed iodine-131 prior to when the cover gas was sampled on August 12, 1959 and that, along with decay, is why I-131 was not detected in it.[65, pp. 44, 48, 51, 71-74] Therefore, it is pertinent to quantitatively assess the extent of venting and flushing of the cover gas and evaluate the effect on the ability to detect and measure I-131 in the sample if the I-131 release to and retention in the cover gas were as Makhijani postulated.

The detailed assessment of all reported operating data regarding cover gas venting and flushing during and after Run 14 is provided in Appendix B. The summary results and conclusions are presented here.

The quantitative assessment of actual operations shows that the venting procedures that occurred on July 12 and 15 removed about 19 percent of the fission product gases that would have been released from the fuels on July 12 and 13 that were postulated by Dr. Makhijani and did not completely flush them out as suggested by Makhijani. This is a significant result. Some 81 percent remained.

Similar evaluations of all venting and flushing operations until the August 12 cover gas sample was obtained show that, as a minimum, 26 percent of the fission gases that may have been in the cover gas on July 12 and 13 remained at the time of sampling, and, most likely, 52%. As discussed in Section XIII, the preponderance of evidence is that, in fact, the majority of fission gases were released on about July 23. In that case, the fraction of original cover gas fission inventory remaining on August 12 is calculated to be 82%. Even when decay is accounted for, there were sufficient gaseous isotopes to be measured in the sample when counted on September 14. Xenon-133 and krypton-85 were both measured. Iodine-131 was looked for and not found.

The quantitative assessment of the relative amounts of I-131, Xe-133, and Kr-85 that would have been in the cover gas sample when counted on September 14, if they were released from the fuels on July 12 and 13 in the amounts postulated by Makhijani, shows that the gamma spectroscopy signal for I-131 would have been 9.4 times the Xe-133 peak and 7.7 times the Kr-85 peak. The sensitivity of the method, derived from measurements on samples of very small concentrations of Xe-133 that were 100 times lower than in the September 14 measurement, show that, had as little as 0.04% of the I-131 in the failed elements been released to the cover gas on July 13 and remained there without reaction with sodium, it would have been measured in the August 12 cover gas sample. For Makhijani's July 13 release date, this corresponds to 1.7 Ci I-131. This was not detected in the sample.

To summarize, if I-131 had been released from the failed fuels to the cover gas on July 12 and 13 in the quantity suggested by Makhijani

- At least 26% of the inventory would still have been present (before decay corrections) when the cover gas was sampled on August 12; this is especially evident in the fact that Xe-133 and Kr-85 were measured in the sample and

- Accounting for decay of all isotopes, the I-131 would have been measured in the gamma spectroscopy scan on September 14. It was looked for and not found. Similar conclusions apply had the fission gases been released from the fuels later on, about July 23. This proves that I-131 was not released to the cover gas.

XV. LESS THAN EIGHTEEN PERCENT OF MAKHIJANI'S POSTULATED I-131 IN THE COVER GAS ON JULY 12 AND 13 WOULD HAVE BEEN VENTED TO THE STACK BY JULY 15

Dr. Makhijani's main hypothesis for release of I-131 to the stack and environment is that the bulk of fission gases were released from fuels failed on July 12 and 13, primarily July13, and that they were completely vented from the core cover gas region to the stack during pressure changes on July 12 and 15 when the bypass valve of the decay storage tanks was presumably inadvertently left open.[65, p. 81] Aside from stack monitoring data that do not support this, a detailed calculation of venting operations in Appendix B (see summary, Section XIV) shows that about 20% of the cover gas was vented during the July 12 and 15 pressure changes.

It is shown in Appendix C that, instead of 1,330 Ci release from venting operations by July 15 as suggested by Makhijani, physically, less than about 236 Ci or 17.7% of the original 1,330 Ci hypothesized by Makhijani (accounting, also, for I-131 decay prior to the July 15 venting) would have been vented and potentially directed to the stack if 1,330 Ci had been present in the core gas originally (ignoring the fact that in the one and one-half day interim from the July 13 excursion to the July 15 pressure venting, all the hypothesized iodine would have reacted with and been removed by sodium – see Section XVIII).

This is not to say that up to 236 Ci actually was released. In fact, complete analysis as described in this report shows that no I-131 was released to the stack. This analysis simply shows that, even if one ignores the fallacies and scientific deficiencies in other aspects of Makhijani's postulated scenarios, only a small amount of his postulated I-131 would have been vented to the stack during the venting operations that he claims to be the basis for the removal of iodine from the cover gas to the stack.

XVI. POTENTIAL MECHANISMS OF FISSION PRODUCT RELEASES FROM FUEL SUPPORT THE OBSERVATIONS OF ABOUT ONE PERCENT RELEASES FROM THE SRE FUEL.

The mechanism of release of fission products from the failed fuels cannot, of course, be definitively established. Three possible mechanisms are:

1. The fission products that had accumulated in the NaK bonding and vapor region of the gap between the fuel slugs and cladding from diffusion processes and recoil during irradiations up to the failure were released with the NaK and the gases in the gas plenum when the cladding ruptured.

2. A portion of the fission products in the uranium that alloyed with the iron in the cladding to form a eutectic mixture that melted and refroze in the sodium were released from it.

3. The destruction of the uranium matrix to a "spongy" form created by multiple transitions through the alpha-beta temperature increased available surface area for fission products to diffuse out at the elevated temperatures.

An assessment of the three potential sources is useful in showing the limits of what may have been released via each mechanism.

The interim report suggested that fission products accumulated in the NaK bond from recoil at the surface of the uranium slugs during irradiation (about 0.035% of fission products in the SRE recoil from the surface) could account for the cesium and iodine found in the sodium.[6, pp. IV-C-6 to IV-C-7] Hart performed a detailed assessment and concluded that, if recoil were the only source of fission products in NaK, it could only account for about 10% of the the Cs-137 in the sodium.[1, pp. 21-22] An evaluation in 1960 of hazards in sodium-graphite reactor systems stated, "Simple ruptures of fuel cladding which release the NaK or sodium bonding alone would generally carry less than 0.1% of the fission product activity of each fuel element into the primary sodium."[66]

Thus, NaK released to the sodium contributed some of the fission product source. If recoil were its only source of accumulated fission products, other mechanisms would be necessary to explain the total release fractions from the fuel into the sodium. In conjunction with mechanism 3 identified above, it is perhaps possible that the cycling between the alpha and beta phases caused additional diffusion from the fuel into the NaK before cladding rupture. This consideration would also hold for krypton and xenon, which were released to the cover gas. Diffusion of xenon and krypton during normal operations is negligible, based on measured diffusion coefficients reported by Barnes.[28] Iodine released from the fuel by these two mechanisms (recoil and diffusion as result of uranium damage from alpha-beta transitions) would have been absorbed into the NaK as sodium iodide and potassium iodide prior to cladding rupture.

An analysis in Section VII, based on the measurement of 800 g uranium-iron eutectic material on a single fuel element R-24, extrapolated to about eight elements assumed to have had eutectic formation, indicated that, if all the fission products in the uranium in the eutectic were released and the average amount of eutectic formed for all the eight elements was also 800 g, it would amount to 0.65% of the total fuel inventory of the thirteen failed elements. This is very comparable to measured fission product fractions in the sodium – see Table VI. However, a number of assumptions are necessary to derive this value. First, it assumes that the fission products associated with the diffusing uranium accompanied it. Evidence is that the eutectic formed mostly by the uranium diffusing into the stainless steel cladding with a lesser amount of iron diffusing into the uranium.[67] This is consistent with the mechanism that would have led to cladding failure, that is, the stainless steel portion melted. Second, it assumes that the eutectic remained molten long enough to release all the fission product contents of the uranium to the sodium by dissolution or, in the case of the noble gases krypton and xenon, by release of gas. Third, it assumes that the same fraction of uranium in each assembly formed the eutectic melt as in the one analyzed element R-24. The average may have been more or less.

The third mechanism identified, damaged matrix release of fission products at high temperature, may be evaluated by considering the known diffusion rates of krypton and xenon in uranium.[28] Barnes et al. have reported the diffusion coefficient of krypton and xenon in uranium as a function of temperature under conditions that represent the fuels failed by cycling through the alpha-beta transition temperature.[28] Because they have nearly the same atomic radii, the diffusion rates are very similar and are represented by a single diffusion coefficient. Measurements were made of diffusion rates of uranium specimens irradiated to 0.0002%, 0.12%, and 0.15%. When the uranium had been cycled between the alpha and beta phases, no change in diffusion rate occurred in the samples irradiated at 0.12% and below. This would best represent the SRE fuels. When the sample irradiated to 0.15 % was transitioned between the alpha-beta phases, the diffusion rate increased rapidly.

The diffusion rate equation calculates the quantity of gas diffused out of the metal for a specified time, temperature and surface area.

These results were applied to the conditions of the SRE reactor. The assumptions were:

- 800 to 900°C (992 to 1,652°F) – the maximum temperature for extended times for most fuels – most were less.
- Exposed surface area of all the slugs in the fuel assemblies summed up.
- 13 days continuous heating following failure by alpha-beta transition on July 13 until reactor shutdown on July 26.

The diffusion parameters for the 0.15% burnup sample that had gone through the alpha-beta transitions were used.

The results show that only between 0.010% and 0.016% of the krypton and xenon would have diffused from the failed elements based on a July 13 failure of all fuels. For the much more probable release date of July 23 (see Section XIII), one-tenth as much would have diffused out over three days. This does not appear to be a significant mode of release.

In considering all the potential mechanisms taken together, the magnitude of about one percent of the failed fuel inventory fission products found in the sodium and cover gas samples are reasonable.

XVII. STACK GAS SAMPLE DATA SHOW THAT LESS THAN ONE CURIE OF I-131 WOULD HAVE BEEN RELEASED IF MAKHIJANI 'S THEORY WERE CORRECT

Dr. Makhijani's postulated main mechanism of proposed loss of iodine from the cover gas is pressure venting operations and that these, combined with decay, would have removed the I-131 so that it would not have been measured in the cover gas sample that was obtained on August 12 and measured on September 14. This is discussed in detail in Section XIV above and shown to be incorrect and not applicable. Secondly, he then contends that an apparent inadvertent opening of the storage tank bypass valve during one of the ventings would have released iodine up the stack. This is purported by Makhijani to have been the main source of release to the environment. Thus, a quantitative assessment of the consequence of the reported venting operations and open bypass valve is important and informative.

The specific occurrences and observations are as follows. On July 15, at 0550, a venting operation of the cover gas was carried out. It was to have been to the holding tanks. At 06:00, the stack gas monitor registered a high reading that indicated a total radioactivity in the gas of $7\times10^{-5}\mu Ci/cm^3$. It continued intermittently high until about 11:00.[6, p. IV-C-24] The stack gas flow was 25,000 cfm. As a very worst case, assuming a continuous high reading for five hours, the maximum total activity released is, thus, calculated to be 15 Ci. If one considers that the July 13 excursion that released fission products that were vented on July 15 represents about 90% of all releases, the quantity becomes 17 Ci. It is actually substantially less as the stack monitor readings fluctuated to lower readings.

If one, for the moment, accepts Makhijani's "probable" release fractions of beta-emitting fission product gases in the cover gas: 45% noble gases and 34.2% I-131 inventory in the failed fuels, the cover gas inventories would have been those tabulated in Table VII (based on July 13 inventories of failed fuel elements when Makhijani postulates the major release occurred decayed to July 15 when the gases were vented two times). See Appendix E for details. The attenuation factors used here are derived in Section XXII.C.2. The attenuation factor is how much the intensity of the beta ray from an isotope is reduced in going through the window of the Geiger-Müller detector. It is dependent on the individual beta ray energy, which is different for each isotope. The relative signals are computed by dividing the activity (curies) by the attenuation factor of the Geiger-Muller detector for the isotope.

Table VII. Stack monitor detection of isotopes in Makhijani's postulated vented fission gases.*

Isotope	Cover gas inventory when vented July 15	Geiger-Müller detector attenuation factor	Relative G-M signal from the isotope
Kr-85	142.5	17	8.38
I-131	1,156	10	116
I-133	1,904	3.77	505
I-135	237.9	2.3	103
Xe-133	3,896	14	278.3
Xe-135	2,624	6.35	413
TOTAL	9,961		1,424

*Based on 45% of noble gases Kr and Xe and 34.2% of iodine isotopes from the failed elements being released to the cover gas and retained there until venting, per Makhijani. July 13 failed fuel inventories were calculated by Daniel & Associates[8] and decayed to the July 15 venting. The July 13 I-131 inventory multiplied by 34.2%, Makhijani's theoretical release, is 1,315 Ci, about the same as Makhijani's 1,330 Ci. Makhijani, however, did not decay the I-131 from his suggested release date of July 13 to the date of his proposed release.

The weighted average attenuation factor for this mix of isotopes in the stack gas would have been 7.00 (= 9,961/1,424). The attenuation factor applied in calculating curies from the detector response was 5.59 (see Section XXII.C.2). Correcting the reported $7\times10^{-5}\mu Ci/cm^3$ to an attenuation factor of 7.00 results in a calculated $8.8\times10^{-5}\mu Ci/cm^3$ and changes the maximum potential total activity release from 17 curies to 21 curies. Of this, the I-131 comprised 11.6% (1,156 Ci I-131/9,961 Ci total), or 2.4 Ci. This would be the outside quantity released from the stack based on the stack monitor response on July 15 if Makhijani's theory were correct (it was not – no I-131 reached the stack).

The percent of the theoretical cover gas inventory released to the stack would be less than 100×(21 Ci measured/9,961 Ci in cover gas) = 0.21%. Based on Makhjhani's theoretical 1,330 Ci I-131 release, the percent measured was 0.18%, because he didn't decay it from date of release from fuels to date of release to the stack. Only 1/500th of his claimed I-131 would have been measured by the stack monitor if releases were continuous for five hours; 99.8 % would have been missed. This is not credible.

Both the actual percent and curies of I-131 released would have been less than these numbers because the stack monitor signal was not at the maximum $8.8\times10^{-5}\mu Ci/cm^3$ continuously for five hours. Considering it to be about half that on average, Makhijani's released fraction would be 1/1000th of less, i.e. more than 99.9% would have been missed. Because a venting operation did not last five hours, but on the order of a minute or less, even this calculated fraction release would actually be decreased dramatically below 1/1000th and the calculated I-131 release would be substantially less than one curie – only a small fraction of a curie.

These conclusions, assuming Dr. Makhijani's postulated releases, are based on the stack monitoring system being functional and accurate. This is addressed and shown to be correct in Section XXII.C.2.

In fact, based on other considerations discussed at length in this report, no iodine was present in the cover gas and the activity in the stack was due entirely to small fractions of Xe-133 and Kr-85 inventories (mostly Xe-133). Based on evaluation of the cover gas sample taken on August 12, assuming the majority of releases occurred on July 13 as suggested by Dr. Makhijani, the calculated percents of these two gases released from the failed fuel elements into the cover gas would have been about 4.3 % of the Xe-133 and 0.062% of the Kr-85 (see Table A5, Appendix A, and Section XIII) and the bulk of the stack activity would have been due to Xe-133, if Makhijani's release dates were correct.

XVIII. ASSESSMENT OF IODINE REMOVAL IN REACTOR COVER GAS AND CONTAINMENT SYSTEM SHOWS THAT IODINE WOULD NOT BE TRANSPORTED TO THE STACK

As documented extensively in previous sections, the iodine was not released from the failed fuel elements as elemental iodine, I_2, and subsequently passed through the sodium to the cover gas. There are a number of barriers to this, starting with the fact that iodine in the metal fuel forms non-volatile metal iodide salt and that even gases in the fuel don't form bubbles that can migrate out at the low burnup concentrations present, followed by the fact that elemental iodine, I_2, would react some with the stainless steel cladding and with bonding NaK to convert to metal iodides, and the fact that elemental iodine reacts immediately with high-temperature sodium to form sodium iodide. The fact that iodine was not released from the fuel elements is substantiated by a number of experiments and observations related to metal fuels with breached cladding in sodium or sodium-potassium-cooled reactors and by measurements in the SRE reactor following the failure of the fuel element claddings. And, assessments of extensive measurements indicate that I-131 was not released to the cover gas.

Nevertheless, even if elemental iodine, I_2, were released to the cover gas as postulated by plaintiffs' expert witness Makhijani, it would not have remained to be purged to the stack gas. It is well known that I_2 reacts with surfaces in a reactor containment vessel to deposit. Dr. Makhijani acknowledges this in referencing a safety analysis study by Atomics International of the consequence of a dropped fuel element[68] and concedes that perhaps as much as 50%, but settles on a probable 22%, could be retained by this mechanism.

However, in the cover gas region above the sodium pool, a much more effective means of removing elemental iodine (I_2) from the gas would have prevailed that would have quantitatively removed it to the sodium pool. The iodine in the gas would have reacted with the sodium by two mechanisms. One would be reaction of gaseous I_2 with sodium vapor atoms that existed above the pool. The other would be by collision of the I_2 gaseous molecules with the liquid sodium pool surface and with sodium condensed on the walls and ceiling above the pool in a reflux process.

Above the hot sodium pool at the minimum temperature of about 541°F (283°C) on July 13, when the major fuel damage is postulated by Makhijani to have occurred, the vapor pressure of the sodium was 1.11×10^{-5} atm and the corresponding concentration of sodium vapor atoms in the gas was 1.5×10^{14} atoms cm^{-3} (atoms per cubic centimeter). If 43.8% the iodine inventory of the failed fuels, including I-127 and I-129 as well as I-131 and I-133, the I-129 predominating in terms of atoms, were released to the cover gas as suggested as most probable by Makhijani (about 0.75 gram) [65, p. 91] the $I_2(g)$ concentration in the about 6,300 L gas would have been about 2.8×10^{14} molecules cm^{-3} (1.8×10^{21} total I_2 molecules). Collisions with sodium vapor atoms would have resulted in reaction to form sodium iodide, NaI. As the sodium gas atoms reacted, they would be replenished from vaporization of the liquid sodium, maintaining a constant concentration while the iodine concentration decreased.

Probably the more predominant process would be reaction of the I_2 gas molecules by collision with the liquid sodium at the surface of the pool. Furthermore, as the sodium vapor refluxed above the pool in contact with cooler surfaces in the gas area, it would condense on the walls, providing even greater surface area for reaction with the iodine. From fundamental kinetic theory of gases, the collision rate with the pool surface (area 88,300 cm^2[5; 10, Figure 2-1]) would have been about 1×10^{23} collisions per second (if no reaction occurred and the concentration in the gas remained constant). Thus, each I_2 molecule would collide with the liquid sodium surface, on average, some 80 times per second. Once an I_2 molecule strikes a sodium surface, it has a very high probability of 'sticking' and reacting, much more effectively than I_2 striking a steel or concrete wall surface that is already acknowledged to be about 50% effective in removing iodine with brief residence time, without regarding chemical reaction with sodium. The removal rate by this process would be limited only by the transfer of the iodine to the sodium surface. This would be enhanced over simple diffusion by convection of the helium gas in the temperature gradients that existed and would be sufficient for quantitative reaction in the time period that the iodine would have been in the containment system above the sodium pool.

This very process was anticipated by A.I. researchers and experimentally characterized. [55, pp.18-22] In the study, Begley showed that 1) the reaction rate of I_2 gas with the sodium pool surface was extremely rapid and 2) the reaction rate was controlled only by the diffusion rate of iodine through the inert cover gas to the pool surface where it reacted instantaneously and that it was sufficient to result in complete reaction in a few hours. The description of the experiment and theoretical analysis is given in Appendix D. Application of the results to the configuration of the SRE indicates that, considering diffusion only and not the convection currents that would, also, contribute nor refluxed sodium on the walls in the cover gas region that would add to reaction, the iodine would have completely reacted with the sodium pool surface in less than 7 hours. For a 2-foot deep cover gas above the sodium pool, which is representative of the SRE, the rate of removal of I_2 from a cover gas of nitrogen at 500°F, again representative temperature, is depicted in Figure 11, which shows that 50 percent is removed in ½ hour, 90 percent is removed in about 3½ hours and complete removal occurs in about 7¾ hours. As described in Appendix D, the fractional removal rate is independent of the starting concentration of I_2. Other effects (convection, sodium on walls and ceiling) would have reduced the time considerably. Also, the modeling was done for I_2 in nitrogen gas; diffusion in helium would be faster. Overall these added effects would cut the time by a factor of at least five (see Appendix D), resulting in 50% removal in 6 minutes, 90% in ¾ hour and complete removal in 1½ hours.

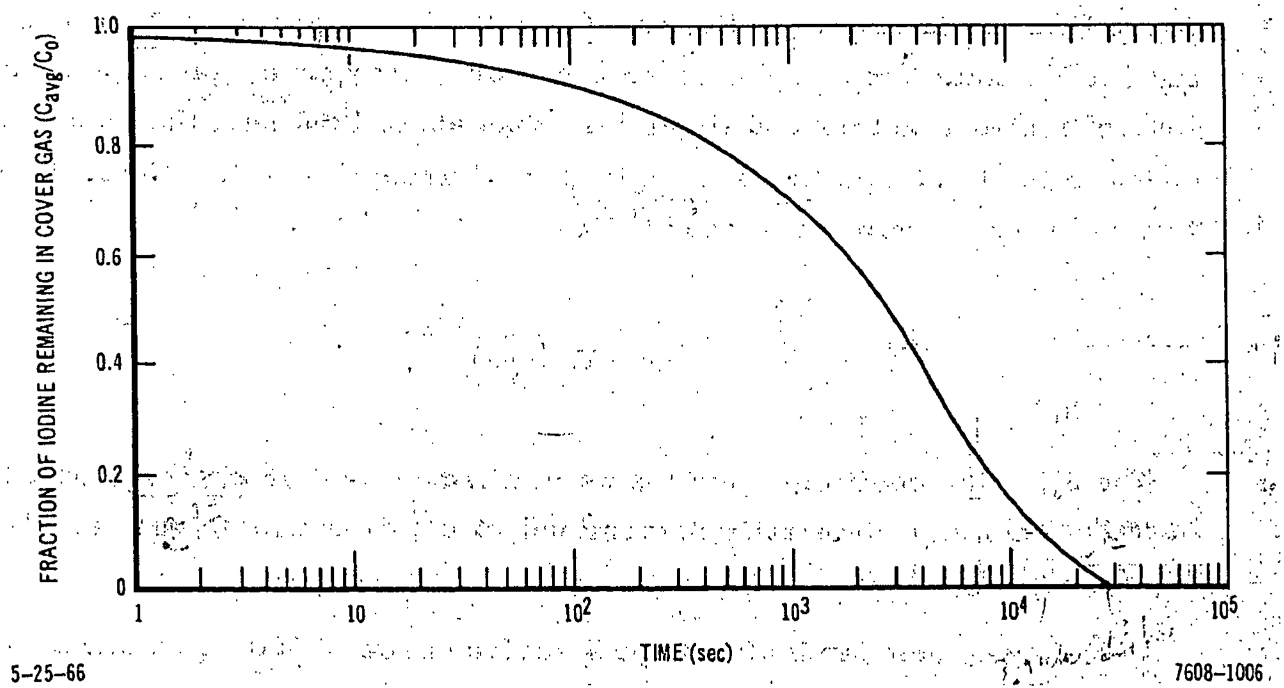


Figure 4. Rate of Iodine Diffusion to Sodium Surface

Figure 11. Removal rate of I_2 from a 2 foot depth of nitrogen above a sodium surface at 500°F.

The most severe fuel pin damage is postulated by Makhijani to have occurred on July 13 during the power excursion and that is when most fission gases would have been released by his theory. That is 36 hours before the next venting of only 20% of the cover gas and less than 18% of Makhijani's hypothetical I-131 fission gas (see Sections XIV and XV). Beyond that additional purging didn't occur until either July 18 or, more likely, July 29. Any hypothetical elemental iodine, I_2, in the cover gas above the sodium would have completely reacted in those interim periods to form sodium iodide, NaI, a salt that would immediately be removed from the cover gas well before any venting.

Over the course of one and one-half days, any I_2 in the cover gas would have had sufficient time to quantitatively react with the hot sodium and be removed as sodium iodide from the cover gas.

Furthermore, and this is very significant, all cover gas that was vented, either to the decay hold tanks or directly to the stack, went either through the sodium fill tank (that had a pool of sodium in the bottom) and then through a sodium vapor trap of baffles and metal mesh that condensed sodium vapors, see Figure 12, or through a line that bypassed the sodium fill tank and through a separate sodium vapor trap.[69] The sodium vapor trap provided a tortuous path that resulted in intimate contact of the gas with an extremely large surface of condensed liquid sodium that would have quantitatively reacted with any I_2 at that point. It is unlikely that elemental iodine could have escaped this only available gas route. The vented helium would have been saturated with sodium vapor and, therefore, would have provided a source of sodium to condense in the trap. Even contact with the very large metal surfaces at temperature above the melting point of sodium would have also resulted in considerable deposition of any I_2 onto the metal surfaces. While this trap design was published in a January 1965 training manual, it is likely to have been the original design. No mention was made of any design changes from the original in the June 1960 tabulation of design modifications.[70] And, vapor traps were shown in the SRE helium system and vent system in 1956 P&I diagrams[71]

XIX. KNOWLEDGE OF METAL FUEL CHEMISTRY IN 1959 WAS INADEQUATE TO EXPLAIN ALL OBSERVATIONS OF THE INCIDENT

The reports assessing the cladding failure incident and the data obtained following it provided thorough description of the conditions during and following the incident, including characterizing the fission product distributions external to the failed fuels.[1, 6] Those evaluations acknowledged the difficulty in not being able to account for the iodine, based on their assumptions that iodine was expected to be volatile as the elemental form, I_2. The interim report, for example, states, "From Table IV-C-4, a release fraction (of fission products, identified in sodium sample 8, analyzed August 8, 1959) on the order of 10^{-4} is indicated. The iodine release fraction falls close to the other isotopes, which indicates that, even though the iodine is very volatile, it did not escape to the cover gas because it undoubtedly combined with sodium as rapidly as it was evolved. No iodine was ever detected in the cover gas samples." [6, p. IV-C-5] No explanation was developed for why the iodine fraction found in the sodium was similar to that of other fission products. Since no analyses were done on the fuel elements, clearly there

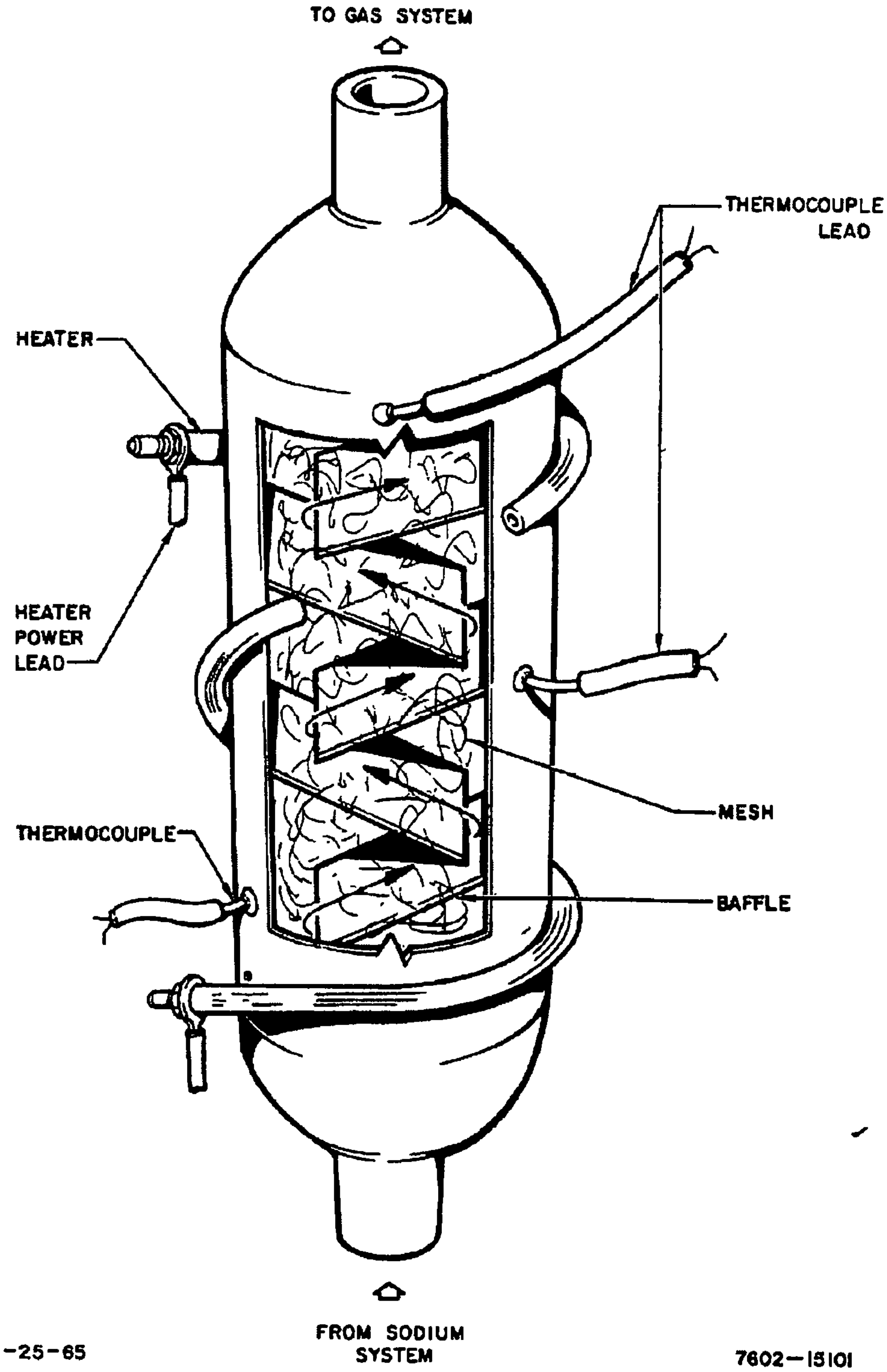


Figure 3-37 Vapor Trap

Figure 12. Sodium vapor trap that vented cover gas went through.[72, p. BNA01134496]

was no recognition that the iodine would have been present as metal iodides in the metal fuel and would not have volatilized.

Hart stated, "However, some anomalies exist. One would expect the release fraction of iodine to be considerably higher than that of cesium rather than one-third of it as actually found," (in reference to relative volatilities),[1, p. 20] and "The iodine release fraction seems unusually small on a volatility basis relative to the other elements. Since neither deposition in the primary system nor escape to the cover gas occurred in detectable quantities, the low sodium value for iodine remains unexplained."1, p. 23] Again, the recognition that the formation of metal iodide(s) in the fuel as providing a complete explanation was not understood as it is today.

It is likely that there was little interest or motivation to explore iodine chemistry in metal fuels at the time (and since) because it was well recognized that any assumed I_2 that may have been released from the fuel would be quantitatively trapped by NaK (sodium-potassium) bonding or by sodium liquid in which the fuels were immersed.

XX. LONG-LIVED IODINE-129 WOULD HAVE ACCOMPANIED IODINE-131 TO THE ENVIRONMENT AND PERSISTED FOR YEARS

When nuclear fuel is irradiated, one of the fission products produced, along with I-131, is its sister isotope iodine-129 (I-129). I-129 has a half-life of 15.7 million years, that is, it is almost stable, though it emits beta particles during its slow decay process. Because of its very long half-life, it persists in the environment for long periods. Iodine combines easily with organic materials in soil. This is known as 'organic fixation' and slows iodine's movement in the environment. Some soil minerals also attach to, or absorb, iodine, which also slow its movement. The long half life of I-129 means that it remains in the environment.[73]

The relative quantities of I-131 and I-129 in the SRE failed fuels on July 13, Makhijani's postulated date of majority release from the fuels, were [8]:

Curies I-129/Curies I-131	9.25×10^{-8}
Grams I-129/Grams I-131	65.0

There would have been 65 times the mass of I-129 released compared with I-131. If 1,330 Ci of I-131 were released to the environment as Makhijani claims as a probable value, there would be 1.2×10^{-4} Ci and 0.7 g I-129 permanently fixed in the environs of SRE. A summary for the values for Makhijani’s postulated minimum, probable, and maximum releases of 366, 1,330, and 2,544 Ci I-131, respectively, is given in Table VIII.

Table VIII. Iodine-129 releases that would have accompanied Makhijani’s postulated July 13 I-131 releases to the environment.

Makhijani’s Postulated I-131 Release, Ci		Associated I-129 Release	
		Curies	Grams
Minimum	366	3.39×10^{-5}	0.19
Probable	1,330	1.23×10^{-4}	0.69
Maximum	2,543	2.35×10^{-4}	1.32

XXI. CONCLUSIONS

The facts discussed above all tie together in demonstrating the following.

1) Experimental and theoretical studies of iodine in metal fuels show that iodine forms uranium and/or cesium iodide salt, a non-volatile chemical form, when it is formed as a fission product in irradiated fuels.

2) Experimental studies of irradiated metal fuels have shown that iodine is not released from heated uranium metal fuel, like that used in the SRE, until after it has melted (which did not occur in SRE) and, then, not as elemental I_2 gas, but as uranium or cesium iodide salt vapor that rapidly condenses at elevated temperatures as it is cooled.

3) Data on operating metal-fueled reactors with failed fuel elements, including melting, show that iodine was not released from the fuels as elemental iodine, I_2, that any release was comparable to other nonvolatile fission products, and that no iodine-131 was ever found in the cover gas or in stack gases. Especially, these observations in the Fermi reactor melted fuel incident bracket the situation of SRE.

4) Experimental data and theoretical modeling of fission gases (i.e., krypton and xenon) in metal fuels show that at less than 1% burnup, they will not agglomerate into bubbles and be released from the fuel. SRE fuel was only burned to 0.1%. The same data and models show that swelling from fission gases must approach 30% before they are released. SRE fuels swelled, not from fission gases, but from the alpha-beta transformations unrelated to fission gas bubble formation-induced swelling to less than 10%.

5) Numerous different experiments showed that elemental iodine (I_2) released into sodium in gas bubbles under violent conditions is retained by the sodium and not released to the cover gas. These experiments more than bracket conditions during the SRE incident in terms residence time and in terms of temperature and rates of mass transport and chemical reaction that result in removal of the iodine.

6) Sodium vapor bubbles formed at the site of assemblies in the SRE would have been broken up by hardware and "goop" at the top of the assemblies as they exited the assemblies. Furthermore, as they traversed up into the cooler pool above the core, the sodium vapor that form the bubbles would have rapidly condensed, causing the bubbles to disappear.

7) Any gaseous iodine (I_2) that would have been in the cover gas (it was not), if released July 12 and 13 as suggested by Makhijani would have been present for at least one and one-half days and some up to five or six days). Experiments and theoretical calculations show that elemental iodine, I_2, would have completely reacted with the sodium pool and vapors in the cover gas region within 1½ hours and been removed as sodium iodide.

8) Any gaseous iodine (I_2) that may have been vented out of the cover gas region to the stack would have had to traverse through a sodium vapor trap of design that would have resulted in complete reaction of the iodine with sodium condensed in the trap.

9) If iodine-131, xenon-133, and krypton-85 had been released to the stack in the quantities suggested by plaintiff expert witness Makhijani, they would have been substantially detected and measured by the stack monitor, even considering design and calibration uncertainties raised by Makhijani. The monitor signal measured would have had to miss more than 99.9 percent of the gases. As discussed in the assessments below of Makhijani's claims, the maximum potential error would have been 36%. The signal would have indicated at least 64%, likely more – 72 to 80% – of his postulated released gas quantity, not 1/1000th of it. Valid stack sampling data show that insignificant amounts of both the noble gases and iodine-131 were released at the time suggested by Makhijani.

10) Krypton and xenon gases were not found in the SRE cover gas in significant quantity as fractions of the fuel inventories, either in the cover gas or the stack from monitoring. A minimum of 26% and a probable value of 52% of the cover gas would have been present after venting and flushing of the cover gas when the cover gas sample was taken, so the cover gas sample would have shown the presence of significant releases had they occurred, but it did not indicate such. The sensitivity of the cover gas sample measurement method would have resulted in detection of as little as 1.7 Ci I-131 from Makhijani's postulated July 13 inventory release.

11) This is consistent with the fact that these noble gases (krypton and xenon) were not significantly released from the failed fuels because a) the fuels were not melted and b) the burnup of 0.1% was insufficient to create gas bubbles that could migrate and be released.

12) Because the fission gases krypton and xenon didn't get released to a great extent, less than about one percent of the failed fuel inventories, iodine, even if present as elemental iodine I_2 gas (it was not), would have similarly been contained in the failed fuels.

13) The small quantity of iodine found in the sodium is comparable to other non-volatile fission products, indicating it was nonvolatile.

14) The absence of I-131 in the cover gas sample, coupled with extensive theoretical and experimental data that iodine in the metal fuel was present as uranium iodide, is consistent with the fact that iodine was retained in the fuels.

XXII. ASSESSMENT OF TESTIMONY OF PLAINTIFFS' EXPERT WITNESS ARJUN MAKHIJANI [65]

A. ASSESSMENT AND CHARACTERIZATION OF INDEPENDENT FACTS

Multiple independent facts provide evidence and lead to the proof and conclusion that no I-131 was released from the SRE stack as a result of the failed fuel elements. All are based on scientific studies or analytical data collected during and after the incident. Sequentially, these include:

1) The fuels with failed cladding did not melt. Literature data for uranium metal fuels show that fission gases at the low burnup of SRE fuels are not released before the

fuel melts. The small amounts of fission gases krypton and xenon found in the cover gas after the cladding failures were primarily released from the fuels as a result of the eutectic melt formed by about one percent of the uranium with iron in the stainless steel cladding at 706°F below the uranium melting point and, perhaps some from uranium that lost some integrity after having cycled through the alpha-beta transition temperature of uranium.

2) Iodine existed in the metal fuel elements in the chemical form of a nonvolatile salt, uranium iodide and, possibly, some cesium iodide, as demonstrated by both experimental studies and theoretical thermodynamic calculations.

3) The very low burnup of the fuel, 0.1%, did not create sufficient fission gas (e.g., xenon and krypton) to form bubbles that could migrate from the fuels. Fission gases were not released in significant fraction from the fuel.

4) Even if iodine were released from the fuels as elemental I_2 (it was not), some or all would have been captured by reaction with the NaK (sodium-potassium) bonding material present between the fuel and cladding and/or by the stainless steel cladding.

5) Only about a few tenths to one percent of the iodine inventory of the failed fuels was found in the sodium, similar to other nonvolatile fission products. Even if iodine were released into the sodium as elemental I_2 (it was not), it would have quantitatively reacted with the sodium to form sodium iodide without release to the cover gas. This was demonstrated conclusively from experiments in which elemental iodine (I_2) was released either by itself or in a bubble of gas in a column of sodium of similar configuration to the SRE sodium pool and fuels. Gas bubbles in the SRE during sodium boiling would have been broken up into smaller bubbles in going through hardware at the top of the assemblies and then collapsed completely as sodium vapors condensed in the cooler pool region above the core (see discussion below, Section XXII.E.2. Bubble sizes in the experiments bracketed this condition.

   It has also been determined by measurement in similar metal-fueled, sodium-cooled reactor incidents, i.e., Fermi and EBR-II, that iodine is not substantially released from failed elements. The Fermi melted fuel incident further showed, by measurement, that no iodine reached the cover gas through the boiling sodium. The iodine did not get released from the sodium in SRE.

6) Even if iodine were released through the sodium pool into the cover gas as elemental I_2 (it was not), it would have quantitatively reacted with sodium in the cover gas environment to form and precipitate nonvolatile sodium iodide during the minimum of one and one-half days prior to any partial venting of the cover gas, and beyond: with sodium vapor above the sodium pool, with sodium refluxed on the walls, and, especially, with the surface of the liquid sodium pool. This is supported by experimental and modeling studies.

7) Even if iodine survived above the sodium pool and was then vented toward the stack (it did not) it would have passed through a sodium vapor trap and been removed by reaction with sodium.

8) If iodine were released through the sodium into the cover gas and remained there as elemental I_2 without reaction (it was not), the cover gas sample taken on August 12 and analyzed on September 14, 1959 would have quantitatively detected and measured it, in spite of various ventings of the cover gas and decay of the I-131 prior to sample collection and analysis. Though I-131 was looked for in the sample, it was not detected. Xenon-133 and krypton-85 were measured but iodine-131, though looked for and not detected, would have been present at greater activity concentration than the detected xenon-133 gas had it been released to the cover gas. At Makhijani's postulated probable releases on or about July 13, the I-131 gamma peak would have been about nine times the observed Xe-133 peak and eight time the Kr-85 peak. I-131would have been detected had as little as 0.04% (1 part out of 2,500) of the failed fuel inventory iodine been released to and remained in the cover gas.

9) Less than 18 percent of I-131 fission gas that was postulated by Makhijani to be present in the core cover gas region would have been vented and potentially available for his postulated transport to the stack as a result of pressure venting operations on July 12 and July 15, 1959. This would correspond to less than 236 Ci I-131 of the 1,330 Ci postulated by Makhijani to have been in the cover gas.

10) Though there were observations of radioactivity in the stack on July 12, it was 1½ hours after the cover gas pressure venting, which would have reached the stack in minutes if not directed to the hold tanks. The stack monitor activity corresponded to operations of the fuel-handling unit.

11) Though there were observations of radioactivity intermittently in the stack at the time of venting of the cover gas to the holdup tanks on July 15, 1959, it was very small. A detailed analysis of the data shows that the quantity of gas that would have bypassed the holdup tanks if the release quantities to the cover gas were as Makhijani postulated was not 100%, which would indicate inadvertent opening of the bypass valves, but would have only amounted to less than 0.1% of the postulated 1,330 Ci I-131 in the cover gas.

12) Stack measurements demonstrated the absence of significant iodine-131 in stack gases at any point during the incident. Stack monitoring was sufficiently sensitive and accurate to detect and measure released gases, in spite of uncertainties identified by Makhijani.

13) Stack measurements of fission gas activity during the July 15, 1959 five hour intermittent excursion compared with known inventories of xenon-133, krypton-85, and iodine-131 in the failed fuel elements at the time of the incident indicate that,

had I-131 also been released as elemental I_2 along with the xenon and krypton in the proportions suggested by Makhijani (it was not), it would have been less than 1 Ci.

Plaintiffs' expert witness Dr. Arjun Makhijani conjectures that about 45% release of the xenon and krypton fission gases and between 9.4 and 65.4%, with a probable value of 34.2%, release of iodine-131 in the failed fuel elements occurred to the stack. In order to support this conjecture, he has had to reject *all* the known science and facts as tabulated above. He starts by stating that the absence of identification of significant iodine at any point in the potential pathway to the stack must mean that the iodine must have been released to the stack. He must, then, explain why the stack measurements did not detect any significant release. The root of the critical scientific error in all of his "analysis" is that he does not consider the fact that the fuel was not analyzed for fission products after the cladding failures and that such analysis would be necessary to complete the material balance that he relies on.

Dr. Makhijani completely ignores items 1, 2, 3, 4, 6, and 7 above. In order to deny the science and experimental facts of item 5, he proposes an incredible mechanism of release of elemental iodine, I_2, through the sodium that is, in fact, proven wrong by experimental testing. He uses qualitative information relevant to items 8, 9,10, and 11 to project quantitative release without a quantitative assessment of the data and uncertainties. Such an assessment would have revealed that no significant quantity of I-131 was present. In order to get the iodine that he suggests escaped through the sodium to be released to the stack, Dr. Makhijani uses the qualitative observation of stack activity during venting in item 9 and the suggestion that the holdup tank bypass valves were inadvertently left open. He proposes that release of most of his postulated iodine inventory in the cover gas occurred at this time (even though the actual venting amounted to less than 20%). To dismiss stack measurements, he claims that the sampling probe was improperly designed and the measuring instrument had insufficient detection limits and was not accurately calibrated. But detailed assessments (see below, Section XXII.C) show that the effects were only minor and insufficient to account for his claim of some 1,000 times greater release of I-131 than would have actually been detected.

Dr. Makhijani's approach to convince his audience of his arguments is to make repeated subjective statements such as, "It is our scientific opinion," or "We derive", or "From our analysis we have concluded that the most probable scientific explanation is" without providing any quantitative analytical basis or documentation. He repeats the same assertions over and over in different sections of his report. For any consideration of his views, it is necessary for him to provide documentation of the basis for his statements so that they may be critically assessed. In such absence, it is shown here how all of these assertions are proved wrong when a complete scientific and quantitative analysis of all the data is done.

It is shown below, in analyzing each of Dr. Makhijani's claims, that in every instance, his general statements and "conclusions" are wrong and are disproved by the facts and data, even liberally allowing the uncertainties that he raises. Most are thoroughly addressed in the text of the independent assessments of the incident, above. Additional issues raised by Makhijani, as just itemized, are addressed in the following.

B. ACCURACY AND DETECTION LIMITS OF COVER GAS SAMPLES

1. Venting Calculations and Cover Gas Measurement of I-131

Makhijani claims that the I-131 that he purports to have been released to the cover gas on about July 13 was not measured in the August cover gas sample that was analyzed on September 14 because of a combination of flushing of the cover gas before sampling and decay of the 8-day half live I-131. He does not mention Xe-133 (5.245-day half-life) and Kr-85, both of which were measured in the sample.

These comments of Makhijani are strictly qualitative based on the understanding that some pressure ventings were reported for July 12 and July 15. He performed no calculations of the degree of venting from the reported data nor of the amounts of decay of the radionuclides to September 14. When these calculations are done, they show that 1) at least 26% of the cover gas remained when sampled, 2) the gamma spectrum signal for I-131 on September 14 in the sample would have exceeded both the Xe-133 and Kr-85 signals in intensity and 3) had as little as 0.04% of the failed fuel inventory of I-131 been in the cover gas on July 13, it would have been detected and measured in the sample. These calculations are described and documented in Section XIV and Appendix B.

Makhijani has no basis for his claim. This assessment of the venting operations also proves that his hypothetical 1,330 Ci in the cover gas was not vented quantitatively to the stack. Only 18% of it would have been possible.

2. Release of Noble Gases from Fuels after the July 29 "Bleed to Negative" of the Cover Gas

Dr. Makhijani, in his expert report[65, p. 72], firmly bases his claim that I-131 was not detected in the August 12 cover gas sample when looked for by gamma spectroscopy because 1) the cover gas was flushed out during pressure venting procedures on July 12, 15, and 29 and 2) the I-131 would have decayed by the time of the gamma scan on September 14 of the sample. He did not explain why, then, was Xe-133, which has a shorter half life than I-131, and Kr-85 were, in fact, measured in the sample. When asked to explain this at his deposition, his response was that all the fission gases, including his postulated I_2, were flushed out on July 15 and 29 and then, subsequently, they continued to be released from the fuels, until shutdown, under conditions that would have resulted in the iodine being trapped in the sodium while the noble gases, krypton and xenon, went to the cover gas.

First, on p. 663 of the third day of deposition, Dr. Makhijani states, "you would likely not be generating much power in the damaged fuel elements." This says that fission gases were not produced in the damaged elements after cladding failure.

Second, he contends in both his report and in his deposition that bleeding the cover gas to negative on July 29 caused the entire cover gas inventory to be removed to the stack and the helium had to be replaced. On page 681 of the deposition, third day, he states, quoting from his report: "In addition to these numerous purges during Run 14, on July 29 the reactor cover gas had been, quote, 'bled down to negative.' That means it was completely purged." That means, according to Dr. Makhijani, that at that point, three days after reactor shutdown, there would have been no krypton, xenon, or iodine in the cover gas that was subsequently sampled on August 12.

Prior to analyzing the consequence of that, it is important to make clear that this statement of 'bleeding to negative' resulting in complete evacuation of the cover gas with accompanying removal of the fission gas inventory is a serious misconception of Dr. Makhijani's. The negative pressure is gauge pressure, that is, pressure in the cover gas region relative to the outside pressure. It is not a vacuum. As described in Appendix B, pp. B5-B6, the operation would not reduce the pressure significantly below ambient pressure so as not to draw air into the cover gas in contact with the sodium. The likely pressure reduction was to about -0.8 psig and the fraction of cover gas released about 18% during the bleed down.

Nevertheless, allowing Dr. Makhijani's theory, any krypton and xenon in the August 12 cover gas sample would have had to be subsequently released from the fuel after July 29, as suggested by him. In Section XVI, diffusion of krypton and xenon out of damaged fuels were modeled during operation of the reactor with fuels at 800 to 900°C (1,472 to 1,652°F), based on measured diffusion coefficients for irradiated uranium metal fuel that had been damaged by the mechanism of cycling through the alpha-beta temperature, exactly as happened in SRE. That showed on the order of 0.01% to 0.02% release over a 13 day period at high temperature. Extending the calculations to 15 days (July 29 to August 12) at 700°F, the temperature at shutdown, the releases of krypton and xenon are calculated to be 0.0003%. This is consistent with measurements in EBR-II run-beyond-cladding-breech tests where only recoil gases from current fissioning were found to be released after initial failure releases (see Section X.C), so none would be released after shutdown.

Dr. Makhijani's attempt to explain the presence and measurement on September 14 of xenon and krypton in the August 12 sample but the absence of iodine does not hold water.

C. ACCURACY OF STACK SAMPLING AND MONITORING SYSTEM

Makhijani challenges the accuracy of the stack sampling and monitoring system on two fronts, without quantifying his assertions. He simply dismissed the monitoring data (p. 48, p. 49 of his report) on the basis of general claims. He claims (1) that the sampling probe in the stack was not properly designed and (2) the Geiger-Muller detector did not have a proper range and was not properly calibrated. On the basis of these points, he contends that the stack monitor would have missed what corresponds to greater than 99.9% of gaseous activity that he claims would have been released. To account for his postulated releases, the stack monitor should have registered for 5 hours an activity concentration in the stack gas of about 6.8×10^{-2} μCi/cm^3 instead of the 7.5×10^{-5} μCi/cm^3 intermittently registered.

1. Stack Sampling Probe Placement and Design

Makhijani (p. 86) cites the ANSI N13.1-1969 standard for sampling in stacks[74] to criticize the design of the SRE stack sampling system as one part of his basis for claiming inability to detect stack activities greater than the approximately 10^{-4} μCi/cm^3 that was observed on July 12 and 15. The first of the criticisms is that the probe in the 3 foot-diameter stack was positioned four stack diameters above the inlet point of the plant gases, rather than the "minimum of five diameters downstream from abrupt changes in flow direction or prominent transitions." This is a relatively small deviation from the recommendation. He further cites a 1991 DOE Tiger Team Assessment that recommends eight stack diameters.

Actual data on the effects of the sample inlet height are available from studies on the Hanford REDOX stack that show only minor effects. At Hanford, a similar situation existed in the 3'9" diameter REDOX stack, but there were two sampling lines, one at 10ft above the inlet (2.7 stack diameters) and one at 40 ft (10.7 stack diameters). The lower probe had a 50 ft long pipe to the monitor and the upper one had only an 11 ft run. McCormack[75] compared the results of measured I-131 from the two probes. The ratio of the 10 ft-to-40 ft results ranged from 0.9 to 1.3, with an average of 1.1±0.1 standard deviation. The lower probe with the longer pipe gave a 10% higher overall result on average. This demonstrates that the magnitude of the effect of probe position is small, on the order of 10%, and, in fact, that, with the lower position, the SRE sample result may have been biased high.

Makhijani's second criticism is that the SRE stack had a single sampling point instead of the five recommended by ANSI N13.1-1969 for stacks with diameters of 30 to 48 inches. That standard was updated in 1999 as ANSI/HPS N13.1-1999.[76] On page 35, the updated standards state: "ANSI N13.1-169 recommended the deployment of multiple nozzles in circular ducts larger than 6-in. diameter or in rectangular ducts with cross-sectional areas greater than 0.5 ft^2. ....The use of these rakes is no longer considered good practice...." ..."In place of multiple point sampling, single point representative sampling should be used, with the requirement that both fluid momentum and contaminant mass are well mixed at the sample extraction location." This suggests that a rake sampling system in the SRE stack would not be recommended today. It also suggests that any effects one way or the other would not be orders of magnitude, i.e., factor of more than a thousand, as would have to be the case for Makhijani's arguments of the sampling having missed the iodine-131 to hold true.

The stack sampling system was adequate for quantitative measurements. The concerns that Makhijani cites are for 10 to 20%-type accuracy and do not result in a factor of 1,000 missed radioactivity as would have to be the case for Makhijani's release theory to be correct. And, based on the Hanford stack measurements, the error may more likely have been to result in artificially high readings of fission gases in the stack and not low.

2. Geiger-Muller Detector Accuracy

In Dr. Makhijani's original report of February 2004[77, p. 21], he relies on a reported ten orders of magnitude range for the Geiger counter in the draft expansion program 1964 safety analysis report[78] to claim insensitivity of the stack monitor alarm to the levels of radioactive gases that he postulates to have been released. He estimated that it was set at a level of emission of about twenty million curies per hour, quite an unreasonable situation – Dr. Makhijani should have realized that no one would design to such an operating limit. This provided a basis for him to explain why the gases were released without detection. However, as he himself evidently later learned, the tabulated range in the draft safety analysis report was, in fact, in error and it was corrected in the final report.[79, p. VIII-28] It no longer supported his argument. Thus, he had to change his line of attack in his final report[65] to defend his suppositions. It is shown in the following that his new argument is without proper basis.

On page 83 of his second report, Makhijani observes that the range of the Geiger-Müller counter used was 1 to 10,000 counts/sec. Then, on p. 85 he comments that the narrow range of 1×10^{-7} to 1×10^{-6} µCi/cc described in 1964 for the expansion program is not consistent with this range. There is no basis for this conclusion. The complete range of the counter is accomplished by means of a switch that selects different scales of narrower ranges up to the maximum. Furthermore, the 1964 report regarding the SRE expansion program that he references for the range 1×10^{-7} to 1×10^{-6} µCi/cc refers specifically to that for a mixture of noble gases, not iodine-131. The report[79], on the same page VIII-28, also gives an instrument range of 1×10^{-11} to 1×10^{-10} µCi/cc for particles. This indicates multiple ranges, although the particulate range may be based on collecting samples on a filter paper for a period of time, which makes specific range comparisons impossible without additional operating information.

It is likely that no specific range was indicated for I-131, simply because it was not considered to be a possible gaseous species ever released from the reactor. That is, monitoring was primarily to detect when the noble gases were being released from the holding tanks. That doesn't mean that I-131 would not have been detected and measured if it were present, if the I-131(plus I-133) signal was in the range of the noble gas signals (and it would have been, see Appendix E, if Makhijani's proposed release fractions occurred).

Finally, one must consider that the design operating range proposed for the expansion project in 1964 may have little bearing on actual operating ranges applied in 1959.

Makhijani criticizes the sampling probe not being isokinetically designed. This would have no effect on the ability to accurately sample gases. Isokinetic design considerations are for sampling particulates. Further, the ANSI N13.1/HPS-1999 standard[76] (p. 35) states, "Studies have shown that isokinetic operation is not a prerequisite for obtaining representative samples."

Makhijani, p. 83, states, "Despite an extensive search, no full record of the stack activity during Run 14 could be found in the provided documentation." The very fact that specific activity concentrations of 1.5×10^{-4} µCi/cc on July 12 and 7×10^{-5} µCi/cc on July 15 were reported has to mean that actual quantitative measurements were in fact taken, in spite of Makhijani's stated inability to locate actual records. The reports would have reflected actual measurements. The lack of actual historical instrument recordings should not mean that the tabulated and reported values are not correct and valid.

On page 84, Makhijani discusses the dependence of the attenuation correction factor of the Geiger-Müller counter on the maximum beta energy of the radionuclides present. He states that the calibration assumed maximum beta energy of 1 MeV, despite the fact that the maximum beta particle energies of the isotopes of interest, I-131, Xe-133, and Kr-85 were somewhat less. He provides a table from an A. I. 1963 report that tabulates the maximum beta energies for a number of isotopes and the corresponding attenuation correction factor for each. A plot of this data in Figure 13 summarizes what he presented. From this, one determines that the attenuation factor for 1 MeV that was apparently used is 5.59. Thus, the errors for the different isotopes can be derived, as shown in the legend of Figure 13. The isotopes I-133, I-135, and Xe-135 are also considered because they would be present along with I-131, Xe-133, and Kr-85. Their attenuation correction factors were calculated from the curve in Figure 13.

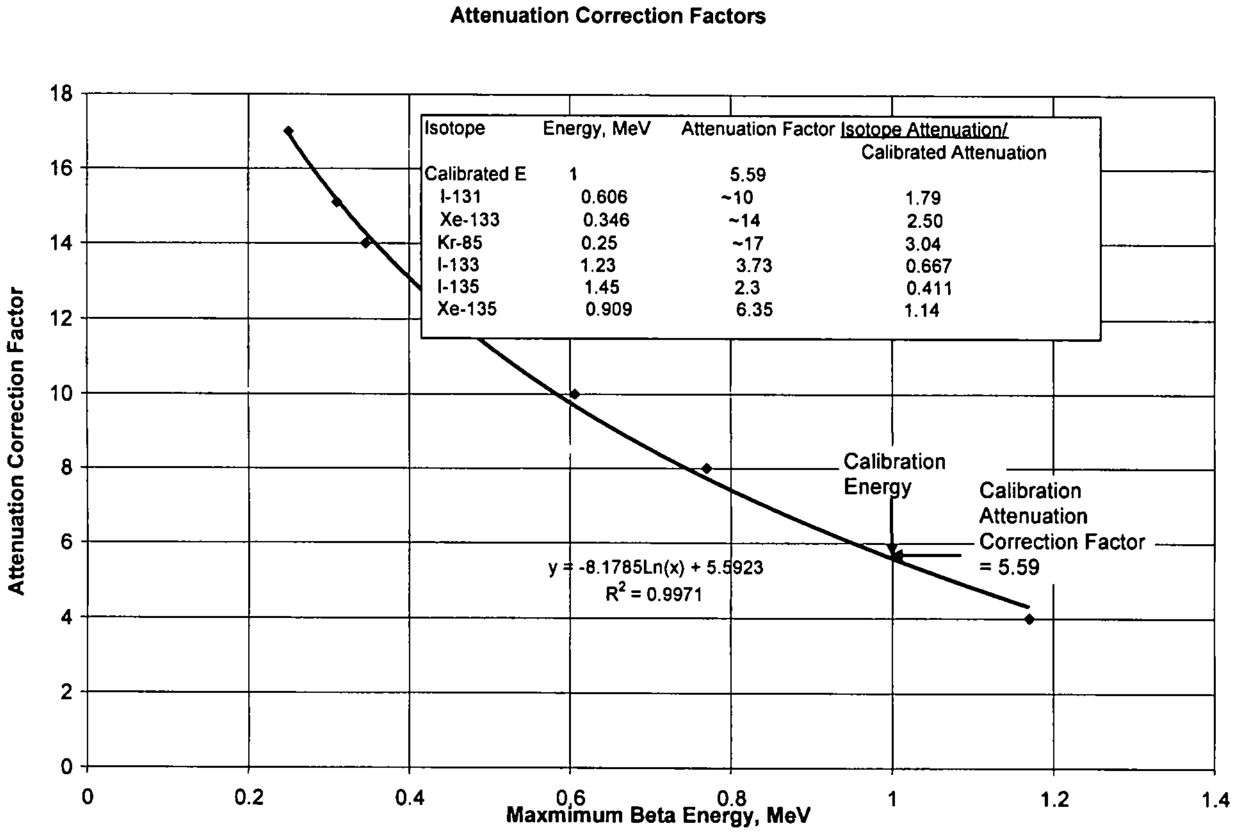


Figure 13. Geiger-Müller attenuation correction factors as function of beta energy, from Makhijani[65, p. 84]

This shows that the error in monitoring I-131 would have been a factor of about 1.8 low (see I-131 line in legend box of Figure 13). For Xe-133 and Kr-85, and Xe-135 the errors would have been factors of 2.5, 3.0, and 1.1 low, respectively, while for I-133 and I-135 it would have been a factors of 1.5 and 2.3 high, respectively.

This means that had about 45% of the Xe-133 and Kr-85 and 34% of the I-131 been released as Makhijani suggests, they would have been substantially detected. Using his suggested release fractions, one derives that the calculated gas mix activity, including I-133, I-135, and Xe-135 contributions, based on the measurements would be 80% of the true activity (i.e., 20% low), if Makhijani is correct in the Geiger-Müller attenuation correction factors. See Appendix E for details. The uncertainty or error of this magnitude would not negate the fact that the signal corresponding to these quantities was never present, but that only the derived total mixed concentration was in error (low) by about a factor of 100/80 = 1.25.

The conclusion is that the stack monitor would have detected the quantities of gaseous activity that Dr. Makhijani suggests was released on July 15 and provided a correct quantitative indication within a factor of less than about 1.5, given all the uncertainties and biases raised by Makhijani. At worst, instead of measuring 100% of the release, it would have measured about 64% (80% GM measurement times 80%, allowing 20% stack probe sampling error – a likely overestimate of probe error that may, in fact, be biased high). Makhijani's claims equate to a measured 0.1% of actual activity. In fact, no iodine was in the stack gases to be detected and only very small fractions of the noble gas source terms in the fuels were released.

D. "BYPASSING" OF HOLDUP TANKS

Makhijani's theory requires that the cover gas vented on July 12 and 15 completely bypassed the holdup tanks where the cover gas was normally routed and somehow vented directly to the stack. This was addressed in Section XVII where it was shown, based on valid stack monitor measurements, that 0.1% leakage past the bypass valves to the stack would have occurred if Makhijani's cover gas inventories were correct. Less than 1curie of his "probable" 1,330 Ci I-131 cover gas inventory (accounting for some decay before "releases") would have been released to the stack.

E. MAKHIJANI'S IODINE "RELEASE" MECHANISMS

1. Basis for Minimum and Maximum Release Fractions from Fuel

Dr. Makhijani has not reviewed or chooses to ignore literature relevant to the release of iodine from the failed metal fuels to estimate the fraction released from the fuels during the cladding failures. See Sections IX and X for relevant data. The most pertinent data is the measured release of iodine from the melted metal fuels in the Fermi reactor incident where between 1 and about 5% of the I-131 and Cs-137 were released from the melted fuel into the sodium under violent conditions and none to the cover gas.

Instead, Makhijani resorts to reports of in-pile experiments, called IP-3 and IP-4, in which thin foils of irradiated uranium metal submerged in 225 cm^3 of sodium at a depth of less than 1½ inches were melted and partially vaporized.[65, pp. 77-78] This series of experiments[80, 81, 82, 55, pp. 37-46] had as their objective simulating conditions of introduction into sodium of iodine and other fission products ejected from failed immersed fuel in order to determine the retention of the fission products by sodium and not of attempting to simulate conditions to reproduce the actual release fractions from failed fuel elements. This included determining the distribution of fission products between the sodium and the helium cover gas. The first report states, "The general objective of the project (7608-4569) is to determine the ability of reactor coolants to retain specific fission products released to the coolant stream as a result of a fuel cladding failure of fuel meltdown. In particular, sodium and organic coolants will be examined as a function of a variety of parameters to determine their ability to retain fission products. The present project is not concerned with studying the detailed fission product release mechanism from fuel during a reactor transient or fuel meltdown, but rather is concerned with the transport of fission products through and release from reactor coolants."[80, p. 4] Thus, Makhijani is misinterpreting the data.

At the start of his assessment, Makhijani acknowledges "In making use of model experiments as a basis for estimating the release of different fission products from the damaged fuel, caution must be used due to the strong dependence of the elements' behavior on the precise geometry and environment utilized in the experiments." Nevertheless, he continues on to use the release fractions of iodine from the melted and partially vaporized uranium foils. He starts by selecting the last two experiments with sodium at 1000°F (vs. 500°F for the first two) because they "were considered to probably have more application toward fast reactor accidents in terms of the simulation conditions," citing Kunkel.[55, p. 45] This reference to being more applicable is in regard to the sodium temperature being more representative for characterizing its retention of fission products, not on what fraction of fission products would be released from the fuel.

In these experiments, a 0.004" (0.1 mm) thick uranium foil was subjected to a sudden burst of neutrons in the KEWB reactor. This caused the temperature to rapidly increase and melt and partially vaporize the uranium foil. The estimated sample temperature achieved was in the range from 2678°F (1470°C) (360°C above melting) to the boiling point, 7052°F (3900°C)*.[80, p. 9] This would have vaporized much of the iodine (including the form of uranium triiodide), cesium, and barium and melted the ruthenium and zirconium whose distributions were characterized. In experiments IP-3 and IP-4, the fraction of iodine released to the sodium ranged from 21 to 72 percent in IP-3 and was 41% in IP-4. Makhijani uses these values to set an "upper" limit of 70 percent released during the SRE fuel cladding failures. (Note, he incorrectly cites 44 percent, not 21 percent for the lower observed value.[65, p.77]) As stated previously, these foil melt/vaporization tests have no bearing to the conditions of the SRE fuel cladding incident. The dynamics of rapid melting, vaporization, and quenching are completely different from swollen fuel that did not melt.

Makhijani goes on to claim that the actual fractional release in SRE was likely greater because of rapid cooling of the foil due to high thermal conductivity and heat capacity of liquid sodium. "It is believed to have been so fast that it froze in radionuclides that would have other wise been released." He cites the first IP-1 experimental report for that statement, which is in reference to why iodine was not found released from the fuel in that experiment. However, that experiment was the only one of the four in which the uranium foil was not pre-irradiated prior to insertion in the KEWB reactor for the melt test in order to accumulated fission products in the uranium. In IP-1, the fission products were formed during the flash irradiation. Since I-131 is mostly formed from parents in a decay chain where the last one before decaying to I-131 (i.e., Te-131) has a half-life for decay to I-131 of 25 minutes, during the few milliseconds of the melting and cooling no I-131 was formed. In that experiment, no I-131 was found in the sodium or cover gas, only some in the frozen fuel that would have formed later from the decaying parents. This is

---

* The boiling point of uranium is currently accepted as 4134°C.

not pertinent to the other experiments where the uranium was pre-irradiated and contained I-131 prior to the start of the experiment. Makhijani is using the above-cited statement completely out of context.

It is notable that the percent of I-131 in the sodium that was released to the cover gas (collected continuously during the experiment by flowing helium over the sample)[82, p.4] was 0.004 to 0.02%. These are the results relevant to the objectives and design of the experiments. The melting and vaporization of uranium, which formed gas bubbles, was accompanied by localized vaporization of sodium. The experiments were designed to "establish the limits of fission product retention. Insofar as possible, in-pile conditions will be chosen to be more pessimistic than any which can be formulated for specific reactor accidents, and thus represent upper limits of hazard conditions which would not be exceeded in any typical reactor situation."[80, p. 30] Thus, these results provide yet another indication that iodine released from actual irradiated fuel under violent conditions will not go to the cover gas. The conditions here melted the uranium and the sodium depth was less than 1½ inches, whereas in the SRE, the sodium pool depth above the fuels was six feet and the fuels did not melt; these experiments more than bracketed the SRE conditions.

To establish a "lower" limit of 20% release, Makhijani uses other completely irrelevant data. While not explained on p. 78 in his report where he lists the value, he elucidated the methodology in his deposition on March 9, 2005 (p. 363ff in deposition transcript [83]). First, he compares the activity (curie) concentrations of I-131 and Cs-137 in sodium during normal operations prior to Run 14 from data given in the interim report, p. IV-C-3, and in Table 5, p. 65 of Makhijani's report. He derives an average ratio of I-131/Cs-137 activity concentrations (ratio of μCi/g Na for each) of 15 from data of a February 6, 1959 sample taken after the January 29 Run 8 shutdown. He then reduces data from the activity concentrations of the two isotopes in a sodium sample taken after the Run 14 releases, correcting for plateout effects, to estimate the percent releases of Cs-137, I-131, and Sr-90 into the sodium from the failed fuels (assuming a July 14 fuel inventory that was partially released), p. 63, Table 4 of his report. He derived 1.39% for Cs-137 and 1.19% for I-131 and notes that, based on the normal operations data, the I-131 percent release should be 15 times the Cs-137 percent release. Thus, 15 times 1.39% yields about 20%, his basis for the lower limit. He also uses the 21% lower value obtained in the IP-3 experiment (which is irrelevant as discussed above) to buttress that result.

This has a number of serious flaws of logic and data and is incorrect. First, it assumes that the relative fraction release of the two isotopes is the same for the fuel failure occurrence as in normal operations. Temperature, fuel matrix condition, and dynamics are all different and one cannot necessarily assume this, although the effects cannot be quantified. More importantly, a significant portion of fission products in the coolant during normal operations were likely produced from tramp uranium impurity in the sodium.[1, p. 6] This would be significant relative to fission products released from the fuel during normal operations where the concentrations in sodium of I-131 and Cs-137 were very small, about 0.017 μCi/g and 0.0007 to 0.003 μCi/g, respectively, but not in the cladding failure situation where the concentrations were 16 and 28 μCi/g (July 26, 1959 basis, not including plateout effects), respectively.

Furthermore, and this is the most important effect (that is, error in Makhijani's logic), the comparison of absolute activity concentrations instead of percent of fuel inventory values in the sodium for the two dates is completely invalid in the type of comparison Makhijani is attempting to make. By comparing activity concentrations in sodium for the two dates, Makhijani is implicitly assuming that the ratios of core inventories of the two isotopes were the same during the fuel failures early in Run 14 as during prior normal irradiations on January 29. This is absolutely not the case. Because of its short half life, 8.04 days, the fission inventory of I-131 depends entirely on the power level of the reactor and the time in reactor up to when steady state is established, which takes about 40 days.[84, p. 5] And, it decays between runs. Cesium-137, on the other hand, with a half-life of 30.17 years, continues to grow with irradiation throughout the operation of the reactor core. Run 14 was at very low power level and of short duration. The fuel inventories for 13 failed elements as of the date of the July 13 excursion were 3,845 Ci I-131 and 2,529 Ci Cs-137, a ratio of 1.52 [8] The February 6 sodium sample was taken following shutdown of Run 8 on January 29 when the curie ratio of I-131 to Cs-137 in the core was 60.2[8]. Correcting this for the 8 days of decay since the January 29 shutdown (a correction in Makhijani's favor that he did not make) results in a ratio in sodium relative to fuel inventories of 30.2. This is uncorrected for plateout effects. Thus, Makhijani's 20% minimum release number must be properly corrected to about (1.52/30.2)×20% = 1.0% as a "lower" bound, still neglecting effects of fission products created from fissioning of tramp uranium in the sodium on the "normal" inventories in sodium, incorporation of which would make the calculated percent release smaller, yet. This (when adjusted for tramp uranium effects – not quantifiable, but could be substantial) is, in reality, not a lower bound, but more a probable value. It is, in fact, quite in line with the few tenths to about one percent inventory of iodine and cesium actually measured in the sodium following the Run 14 fuel failures and supports the model of iodine being released from the fuel as a non-volatile species, i.e. UI_3 and/or CsI, in the same magnitude, percentage-wise, as other non-volatiles.

In summary, Makhijani's statements of bounding maximum and minimum percent release of I-131 from the failed fuels are entirely without basis and merit. He misapplies IP-3 and IP-4 data to irrelevant conditions for the upper limit and he performs an incorrect nuclear physics and mathematical treatment of the sodium data for the lower limit. Neither result is correct. While his calculation of so-called "minimum" release fraction is in error, when corrected, it actually provides a rough estimate of probable (actual) release fraction of I-131 from the fuel into the sodium of a few tenths to about one percent.

2. Bubbles in Sodium

Dr. Makhijani argues that sodium vapor bubbles formed at the site of the ruptured cladding and persisted as large bubbles after they escaped from the fuel assembly region and traversed on up to the cover gas through the six feet of cooler sodium pool above the core, carrying fission product krypton, xenon, and elemental iodine gases with them. He further dismisses data of relevant experiments in which elemental I_2 was released into sodium, either alone or mixed with other gases in a bubble that showed negligible I_2 escaped from the sodium (see Section XII for description of these experiments). His dismissal is based on the argument that bubbles in the experiments would have been small while those in SRE were large, thus, enhancing the reaction of I_2 with sodium in the bubble experiments relative to in the SRE, all the while ignoring the extremely large effects of the higher temperature in SRE sodium on the rates of processes that would lead to much enhanced reaction of I_2 with sodium relative to the experiments. The temperature effect is discussed in Section XII. The bubble size is discussed below.

Once sodium vapor bubbles that Dr. Makhijani postulates to contain the elemental iodine (I_2) and noble gases exited the fuel channels where the sodium was boiling, they entered the bulk region of sodium above the fuel that was dramatically cooler. The channel exit temperature during the July 13 excursion was about 755°F (402°C) and the temperature towards the top of the sodium pool decreased to about 542°F vs. the boiling temperature of 1621°F (883°)[6, pp. III-11 and II-12]. This would have created a tendency for the sodium vapor bubbles to rapidly collapse, that is, for the sodium vapor to condense and go away, leaving Makhijani's postulated small fractions of associated fission gases to be in the sodium vapor bubbles to remain in extremely small residual bubbles. Then, the postulated I_2 would be in intimate contact with the hot liquid sodium and would have instantly reacted with it (if not with the sodium vapor previously). This is quite a different situation from where the entire column of sodium is heated to the boiling temperature that could sustain sodium vapor bubbles.

Dr. Makhijani, in his deposition [77, p. 363] acknowledged that the sodium plenum above the fuels was not at boiling temperature. He cannot dispute that sodium vapor atoms formed in the liquid at the boiling temperature of sodium would condense in the cooler sodium.

A second very significant aspect relevant to sodium bubble dynamics is the design of the fuel assemblies in the moderator cans. In Figure 1 is the picture of a model of a fuel assembly being inserted into a moderator assembly. It can be seen that the space between the assembly and the moderator can wall is very small. The gap from the fuel rods is about 0.08 inch. Free space within the assembly of six rods surrounding a seventh in the center is confined – see Figure 1 depiction of seven-rod element. The sodium flow was not over a smooth surface with full opening at the top. First, the individual fuel rods were wrapped with wire to separate them. This would tend to break up bubbles as they traversed through the assembly. Then, at the top of the assembly was the hanger hardware. Associated with it were two items that would have served to completely break up any large bubbles as they were released into the cooler sodium pool above. One was a hanger tube "designed to serve as a hold-down for the moderator cans if they should, for any reason, tend to rise in the core. The tube had twelve 3/4-inch diameter drain holes cut in the wall and six 5/8-inch diameter holes in the end plate to insure sodium drainage when the element is lifted out."[6, p. II-A-7] The second was an orifice in place[85] to control the flow of sodium individually (in combination with an orifice plate at the bottom of the assembly) in order to maintain uniform temperature across the core with assemblies that had different power densities. The effect of these two structures at the top of the assembly is evident in the fact that they were clogged with "goo" on element R-55, one of the failed elements that was removed and examined following Run 14.[85, 86, p. 4] This shows that substantial hardware trapped particles and would, likewise, have interfered with and broken up bubbles. The "goo", itself, that accumulated from the tetralin breakdown products (and eventually caused the overheating that resulted in boiling) would have provided an even more restrictive path for the bubbles (as it did for liquid sodium flow) and resulted in additional bubble breakup.

A second occurrence of plugging of the drain holes was the cause of the wash cell incident following the shutdown of the reactor after Run 13.[87] The fuel element from channel R-56 was removed and found to be dirty, though in otherwise good shape. While washing it in the wash cell, a sodium-water reaction occurred that resulted in an explosion that severed the shield plug and hanger tube 18 feet into the air. It was determined that sodium had not flowed out of the drain tube above the assembly (described above) because it had been clogged by hydrocarbon deposits formed from decomposition of tetralin.

Any sodium vapor bubbles that formed from boiling within the assemblies would have been dispersed by these various constrictions as small bubbles into the cooler sodium pool above the assemblies, thus, enhancing both the collapse of the bubbles there in the decreasing temperature gradient and reaction of any postulated elemental iodine (I_2) in them reacting with the sodium. Makhijani's theory of large sodium vapor bubbles escaping through the sodium plenum above the core cannot be defended in the face of this breakup mechanism and the condensation of sodium vapor in the bubbles as they contacted the cooler sodium pool region.

Makhijani's postulated model of the iodine being transported through the sodium pool to the cover gas is not credible.

F. MAKHIJANI'S ESTIMATES OF IODINE PLATEOUT IN COVER GAS REGION

Dr. Makhijani cites a number of A.I. safety analysis evaluations regarding hypothetical iodine releases and plateouts and information of removal in stack condensate of iodine evolved from a fuel dissolver at Hanford to "derive" his estimates of the fraction of iodine that he postulates to be in the SRE cover gas that would plate out.[65, p. 82] On this basis, he estimates that between 5 and 50% of iodine postulated to be in the cover gas plated out prior to release to the stack, with a probable value of 22%. His evaluations are flawed on a number of fronts.

- He misapplies SRE estimates to conditions that are not pertinent to the hypothetical iodine in the cover gas.
- He misquotes some of the SRE plateout values provided – he leaves out sequential plateout steps.
- He ignores greater plateout values mentioned in the safety analysis reports and, even in his own report, than the maximum 50% that he selects.
- The Hanford data is misinterpreted and, in any event, is completely irrelevant to the SRE reactor.
- Estimated plateouts in all the A.I. safety analyses are guesses and, in fact, are less than that. They are assumptions. The assumed fractions plated out are intentionally underestimated for conservative assessments of safety impacts.

- He incorrectly applies assumptions used in safety analyses that are, in fact, minimum plateout values as maximum plateout values.

- The safety analyses that he cites are for sudden releases, while the SRE gas resided in the cover gas region above the sodium pool for days before release by venting.

- He ignores reactions of hypothetical iodine in the cover gas with sodium while it resides there for days before any release to the holdup decay tanks or to the stack, both via the sodium fill tank and sodium vapor trap.

These are addressed point-by-point in the following discussion.

Makhijani's first citation is a fuel handling accident safety analysis assessment that assumes that 50% of iodine released from a fuel element dropped on the reactor high bay floor and oxidized gets transported to the environment through the building ventilation, i.e., 50% is plated out on walls of the high bay area.[88] He applies this to state that a maximum of 50% of iodine in the cover gas region above the sodium pool would plate out prior to reaching the stack. First, as stated above, the 50% value is an assumption and, for the safety analysis would correspond to a minimum value. Second, the conditions are not pertinent at all to iodine in cover gas that is exposed to sodium and then released to the stack via the sodium fill tank and through the sodium vapor trap.

The second A.I. citation by Makhijani is the analysis of the consequences of a core melt accident in the SRE power expansion program report.[79, p. C-3] The safety assessment assumes conservative factors of 100% release of iodine in the fuel to the sodium, 0.1% release of iodine in sodium to the cover gas, and 0.02% release of iodine in the sodium to the high bay, the latter of which corresponds to 20% of the iodine in the cover gas. This value assumes plating of iodine in the core cover gas cavity or between the cavity and a relatively cool path, such as around the top shield plugs, which the gas would traverse to enter the reactor building. This states that 80% of iodine in the cover gas plates out before reaching the high bay. To this, one must then apply the 50% plateout on the high bay walls that is assumed from the fuel dropping accident, for a total of 90% plateout of the cover gas before release to the environment.

Makhijani not only ignores the 80% plateout assumption of cover gas iodine that is much more representative of the fuel failure in the reactor scenario, but he neglects the additional 50% plateout in external surface exposures such as piping and vessels between the reactor and the stack. This 90% plateout is not considered a maximum, but by the nature of safety analyses, would be a minimum assumption by the safely analysts. That is, substantially higher plateouts are likely but are not credited in the safety analysis. Never in a safety analysis is an optimistic scenario assumed but always the most pessimistic one in order to conservatively bracket all possible conditions. Thus, the derived safety and environmental consequences are conservatively the worst so that the plant can be designed to assure safety.

Note as an aside, also, the assumption of 0.1% release of iodine from the sodium to the cover gas in this safety analysis, which, again, is an overestimate by about a factor of ten of what was considered at the time in 1964.

Makhijani ignores this second report and relies on an informal letter report that responded to a query about dose consequences with a *specified* release of 5% of iodine to the cover gas in a core meltdown.[89] In the letter, Hale states at the outset that several reports have been written indicating iodine release through a sodium environment is very small, of the order of 0.01%. Nevertheless, he performed a radiological analysis assuming 5% release from the sodium and 2.5% release from the building, based on an assumption that one-half of the iodine is plated out over the leakage paths from the reactor and in the reactor building. There is no basis provided for the assumption. Hale also went on to perform a dose calculation based on a more realistic assumption of 0.02% release from the sodium.

Makhijani's assumption of 50% maximum plateout, based on review of the safety assessments, is without merit. The value he should have selected as a *minimum* plateout is that of the core meltdown scenario in the SRE power expansion official report, i.e., 80% coming out of the cover gas and then 50% of the 20% iodine released from the cover gas plating out external to the cover gas region as provided in the fuel dropping assessment, that is, 90% minimum plateout overall. It should be considered a minimum value because the safety analyses are always based on the worst premises, not those leading to least negative impact. The purpose of safety analyses is to bracket the worst possible assumed condition. Selecting a maximum probable plateout is opposite to this purpose.

Finally, Makhijani establishes his minimum of 5% plateout assumption based on data reported for operation of the stack for diluted fuel dissolver off gas at Hanford. In this case, he, first, misrepresents the Hanford data in terms of what it means. Second, it has absolutely no relevance to conditions in the SRE.

Makhijani, p. 82 of his report, states, "The 1993 Hanford Environmental Dose Reconstruction (HEDR) project cited three studies from 1946 regarding the percentage of iodine lost to the stack following the dissolution of irradiated fuel elements.... In two of the references a loss of 5 percent of the iodine was reported, while the final reference listed zero losses to plating during the summer." The HEDR report that he references[90, pp. 4.26-4.27] tabulates data from two reports, and they likely draw from the same information. The first is a letter from Smith to Tilley that mentions 5% removal of the total processed iodine in stack condensate.[91] The second is a letter from Work that also indicates 5% removal of total processed iodine in stack condensate but that there may be no condensate in the summer.[92] Work references a memo form Tilley to Peterson for his information. The 5% removal of iodine in the stack was in stack drainage at the bottom of the stack. The third reference[93] provides the evaluation that in the summer there was no condensate. Thus, the conclusions of no scrubbing in the summer.

It should be noted that the 5% refers to the percent of iodine processed in the fuel dissolvers, not of that released from the dissolver and transported to the stack, which, in the early days when the 5% in stack condensate was measured, was 50% of inventory[94] and, later, at higher burnup, 85% of inventory. The percent of iodine in the gas recovered in condensate was 10%, not 5%. Nevertheless, these Hanford stack drainage data have absolutely no relationship to what would happen to hypothesized iodine in the SRE environment, both above the cover gas and during transport from the cover gas to the stack or the high bay and its exhaust. Makhijani's selection of the (incorrect in itself) 5% removal as a lower limit, which he uses in deriving a "probable" removal fraction, is completely without scientific merit.

In addition to his incorrect assumptions and selective use of data, Makhijani applies incorrect statistical averaging to his selected minimum and maximum values to derive a "probable" value. First of all, with two limiting values so widely apart, one cannot make a statistical determination of the distribution of values and, therefore, of a probable value. If one were to attempt to, the best approach is to simply take the average, which approximates a normal distribution. Makhijani selects the midpoint of the arithmetic average (27.5 – one-half of the sum of the two) and the geometric mean (15.8 – the square root of the product of the two). This decreases the "probable" value from the straight arithmetic average that would be most appropriate.

In summary, Makhijani's treatment of iodine plateout is flawed. He applies assumed minimum plateout values as maximum values. He ignores the more pertinent 80% plateout for iodine transport from the cover gas and selects only the 50% value assumed for gas released in the high bay area. He incorrectly applies irrelevant Hanford stack condensate data as a minimum value and even errs by a factor of two in its value. He applies an incorrect methodology to derive a probable value from his selected minimum and maximum values that decreases the result from the more correct arithmetic average. And, finally, he ignores the fact that his postulated iodine in the cover gas would have been in contact with sodium for at least one and one-half days before any venting and then would have been vented through a sodium vapor trap (Figure 12) where any iodine would have been removed.

In order to correctly develop minimum, maximum, and probable plateout values, using his methodology of utilizing assumed safety analyses values, he should have used 90% as the minimum value, 100% as the maximum value, and the average of these, 95%, as a probable value. Even this would not be defensible because the minimum values are assumed small for safety analyses purposes and are not, in fact, realistic. More significantly, the chemical environments to which any hypothetical iodine in the cover gas would have been exposed would have resulted in complete reaction with and removal by sodium (see Section XVIII).

G. MAHKIJANI'S "INDEPENDENT" REVIEW BY SCHUMACHER

Dr. Makhijani purports to validate his theories and findings by obtaining an "independent" review by Dipl. Phys. Otto Schumacher.[95] Schumacher's review is superficial and not substantiated. His analysis demonstrates a lack of understanding of

- the differences in chemistry between metal and oxide fuels
- the influence on burnup on the release behavior of the noble gases from metal fuels
- the quantitative amount of cover gas venting that occurred during the pressure reliefs of Run 14.

Schumacher's review cannot be considered in any sense of the word as an independent "peer" review. A few of his comments are reviewed here to demonstrate the insufficiency of his review in substantiating Makhijani's theories.

In his conclusions section 2.2, he states,

> "Based on a rather crude assessment of noble gases found in the cover gas volume in /2/ (reference 1 of this report) it was estimated in /2/ that some 0.1% of core inventory had been released. This estimate must be regarded as completely useless since at least two times (on July 12 and July 15) the cover gas had been vented in the meantime /8/ (reference 6 of this report)."

In fact, Hart accounted for the venting and in this report, Appendix A, detailed assessments of the venting are made that show that less than 20% of the fission gases in the cover gas were vented on July 12 and 15. As with Makhijani, Schumacher avoids details that would disprove his assertions.

Continuing in his conclusions, Schumacher states,

> "Regarding the Cs-137 findings in the sodium, the lack of substantial amounts of iodine was also noticed in /2/ but was not explained. This inconsistency is even more obvious, when the Sr-90 values are taken into account as well. From /5/ it is known that amounts in the order of those, measured in the sodium, are only released in case of melting of fuel. For these cases nearly the complete inventory of noble gases and iodine is released and large parts of cesium. Thus as compared to Sr-90, a lack of Cs-137 must be recognized."

His reference /5/ is regarding the SNR-300 reactor, which was a breeder reactor design fueled with mixed oxide (MOX) fuel of uranium oxide and plutonium oxide. The reactor never operated. As discussed extensively in this report, a thorough literature review shows that melting oxide fuels do not represent non-melting metal fuels of the SRE. It is shown from experiments that noble gases are not released from the metal fuels at the low burnup of SRE fuels and that iodine is not in the form of volatile elemental I_2 in metal fuels but is in oxide fuels.

Schumacher goes on:

> "... in view of the temperatures that were achieved during the excursion there is no reason recognizable why gaseous fission products should not have been released from the fuel pins to a large extent. This holds especially true for the iodine that was produced during runs 12 and 13. During these runs, thermal power of the reactor was 15 and 14 MW, averaged over the operation time with longer phases of nominal power of 20 MW. At this higher temperature as compared to 1 MW power of run 14 volatile fission products must be supposed to diffuse from the metallic fuel to the fission gas plenum of the fuel pins."

This statement is fraught with weaknesses and flaws. First, Schumacher clearly states that his observations are suppositions. He provides no substantive basis for the claims. Once again, he does not recognize the literature that shows that the noble fission gases, krypton and xenon, would not be released from the SRE metal fuels at the low 0.1% burnup and he does not recognize literature and theoretical demonstration that iodine exists as non-volatile metal iodides in the metal fuels. Additionally, the temperature of the failed fuels in Run 14 exceeded the temperature in Runs 12 and 13, even though the latter were at higher power, because the sodium flow was unrestricted in them. Finally, even if his statement were true about the iodine having diffused from the fuel in the form of I_2 to the fission gas plenum during runs 12 and 13 (it is not), he ignores the fact that the iodine would have reacted quantitatively with the NaK bonding material in the gap and converted to sodium iodide and potassium iodide before run 14.

He then states,

> "From /5/ and /6/ (again references to the SNR-300 oxide-fueled reactor) it is known that nearly all of the iodine will be released from the fuel pins as elementary iodine, which is supposed to react quickly with sodium."

Once again, his lack of understanding of iodine chemistry in metal fuels and of the chemistry being completely different from that in oxide fuels is displayed. Even more, his comments about iodine release from the oxide fuels are in reference to melted fuels, oxide fuels melt at much higher temperatures than uranium metal, and SRE metal fuels did not melt.

XXIII. SUMMARY COMMENTS

The analysis in this report documents that no iodine-131 was released from the SRE stack during or as a result of the July 1959 incident. No elemental iodine, I_2, was released from the fuels. I reserve the right to comment on any opinions offered by plaintiff experts at trial.

May 26, 2005
Date

Jerry D. Christian
Signature, Jerry D. Christian

415-856-7100

XXIV. REFERENCES

1. R. S. Hart, "Distribution of Fission Product Contamination in the SRE," NAA-SR-6890 (March 1962). BNA08401400.
2. Nucleonics Reactor File No. 3, Reactors on the Line, Sodium Reactor Experiment, Nucleonics, Vol 15, December 1957.
3. C. Starr, "Sodium-Graphite Reactor," Proceedings Conference on Peaceful Uses of Atomic Energy, May 13-17, 1957, Japan.
4. "Sodium-Graphite Reactors, Sodium Reactor Experiment," pp. 72-90 in *Reactor Technology, Selected Reviews – 1964*, Edited by L. E. Link.
5. "Sodium Reactor Experiment for the United States Atomic Energy Commission" booklet on technical information of the Sodium Reactor Experiment by Atomics International, INEEL Technical Library call number TK9203 S6S679. BNA05076996.
6. A. A. Jarrett, General Editor; R. L. Ashley et al., "SRE Fuel Element Damage, an Interim Report," NAS-SR-4488 (November 15, 1959). BNA03439250.
7. M. Benedict, T. H. Pigford, and H. W. Levi, *Nuclear Chemical Engineering*, Second Edition, McGraw-Hill Book Company, New York, 1981.
8. Daniel & Associates, Inc., 136 Parker Road, Crescent City, Florida 32112-4736.
9. R. R. Durand et al., "SRE Standard Operating Procedures," NAA-SR-MEMO-5326 (June 27, 1960). BNA00457851.
10. J. O. Nicholson et al., "Sodium Reactor Training Manual," NAA-SR-6895. BNA01134270.
11. R. L. Ashley et al., "Evaluation of the Atomics International Nuclear Development Field Laboratory as a Location for Reactor Facilities," R. L. Ashley, General Editor, NAA-SR-7300 Special (May 25, 1962). BNA08401054.
12. R. D. Burch, "SRE Facility Survey," Atomics International letter to Sodium Reactor Committee, February 28. 1963. BNA02008329.
13. Figure A-IV-3, p. A-IV-3c, Gas Services, P and I Diagram (Sheet 3 of 3; 9693-70020), June 27, 1960. No BNA number provided. This figure falls between Figure A-IV-3, p. A-IV-3b, Sheet 2 of 3; 9693-70019, BNA00458114 and Figure A-IV-B, p. A-IV-4, BNA00458115.
14. K. H. Campbell and G. E. Deegan, "Manual of Standard Operating Procedures for the Sodium Reactor Experiment," NAA-SR-1736 (date illegible). BNA0397732. Same as BNA07879504, December 26, 1956.

15. G. Friedlander, J. W. Kennedy, and J. M. Miller, *Nuclear and Radiochemistsry,* Second Edition, J. Wiley & Sons, New York (1966), p. 314.

16. H. Kleykamp, "The Chemical State of the Fission Products in Oxide Fuels," J. Nuclear Materials, **131**, 221-246 (1985).

17. S. Sunder, "The Relationship Between Molybdenum Oxidation and Iodine Volatility in Nuclear Fuel," Nuclear Technology, 144, 259-273 (2003).

18. A. W. Castleman, Jr., "The Chemical and Physical Behavior of Released Fission Products," BNL-6415 (1962) and The Third Conference on Nuclear Reactor Chemistry held in Gatlinburg, Tennessee, October 9-11, 1962, UKSAEC Report TID-7641 (1963).

19. A. W. Castleman, Jr. and I. N. Tang, "Vaporization of Fission Products from Irraditated Uranium. II. Some Observations on the Chemical Behavior of Fission Products Iodine and Cesium," BNL 13651 (1969).

20. J. L. Ballif, "Metallurgical Aspects of SRE Fuel Element Damage Episode," NAA-SP-45115 (1961). BNA08399718.

21. G. W. Parker and C. J. Barton, "Fission-Product Release," Chapter 18 in The Technology of Nuclear Reactor Safety, Volume 2, Reactor Materials and Engineering, The M.I.T. Press (1973), pp. 525-618.

22. A. Roine, Outokumpo HSC Chemistry® for Windows, Chemical Reaction and Equilibrium Software with Extensive Thermochemical Database, Version 5.1, December 9, 2002, Outokumpo Research Oy Information Service, P O Box 60, FIN-28101 Pori, Finland; available from ESM Software, Hamilton, Ohio.

23. T. M. Adams, H. B. Peacock, A. J. Duncan, and M. E. Hodges, "The Melt-Dilute Treatment Technology Offgas Development Status Report," WSRC-TR-00145 (April 199).

24. R. L. Ashley et al, "SRE Fuel Element Damage Final Report," NAA-SR-4488 (1961). BNA03430428.

25. John Krsul, Idaho Falls, Idaho, personal communication to J. D. Christian, September 23, 2004.

26. L. C. Walters, B. R. Seidel, and J. H. Kittel, "Performance of Metallic Fuels and Blankets in Liquid-Metal Fast Breeder Reactors," Nuclear Technology, **65**, 179-231 (1984).

27. G. L. Hofman, L. C. Walters, and T. H. Bauer, "Metallic Fast Reactor Fuels," Progress in Nuclear Energy, **31**, 83-110 (1999).

28. R. S. Barnes, A. T. Churchman, G. C. Curtis, V. W. Eldred, J. A. Enderby, A. J. E. Forman, O. S. Plail, S. F. Pugh, G. N. Walton and L. M. Wyatt, "Swelling and Inert Gas Diffusion in

Irradiated Uranium," Proc. of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy Held in Geneva 1 September – 13 September 1958, Vol. 5, Properties of Reactor Materials (1958).

29. F. G. Foote, "Anomalous Swelling of Mark IA Fuel," Annual Progress Report for 1966 – Metallurgy Division, ANL-7299 (1967), pp. 102-113.

30. W. N. Beck, R. J. Fousek, and J. H. Kittel, "The Irradiation Behavior of High-Burnup Uranium-Plutonium Alloy Prototype Fuel Elements," ANL-7388 (1968).

31. R. S. Barnes, "A Theory of Swelling and Gas Release for Reactor Materials," J. Nuclear Materials, **11**,(2), 135-148 (1964).

32. L. C. Walters, Argonne National Laboratory, retired, Idaho Falls, Idaho, personal communication to J. D. Christian.

33. W. N. Beck and R. J. Fousek, "Fission-Gas Release and Thermal Conductivity Measurements on U-5 wt% Fs Irradiated In CP-5," Trans. Amer. Nuclear Soc., **12**, 78-79 (1969).

34. N. R. Chellew and M. Ader, "The Melt Refining of Irradiated Uranium: Application to EBR-II Fast Reactor Fuel. XI. Behavior of Iodine in Melt Refining," Nucl. Sci. and Engineering, **9**, 82-86 (1961).

35. P. R. Monson, H. W. Randolph, P. R. McClure, M. T. Leonard, P. G. Ellison, M. L. Hyder, and D. L. Hagrman, "Fission Product Release Phenomena during Core Melt Accidents in Metal Fueled Heavy Water Reactors," EDTV-MS-90-15 (1990); WSRC-MS-90-15.

36. "October 5, 1966 Fuel Damage Incident at the Enrico Fermi Atomic Power Plant – Status as of February 24, 1967," NP-16750 (1967).

37. "Report on the Fuel Melting Incident in the Enrico Fermi Atomic Power Plant on October 5, 1966," APDA-233 (December 15, 1968).

38. RadDecay for Windows, version 1.13, Grove Engineering (1995-1996).

39. A. W. Castleman, "LMFBR Safety, I. Fission-Product Behavior in Sodium," Nuclear Safety, **11**, 379-390 (1970).

40. A. W. Castleman, Jr., et al., "Fission Product Behavior in Sodium Systems," in *Alkali Metal Coolants,* Symposium Proceedings, Vienna, 1966, pp. 729-742, International Atomic Energy Agency, Vienna, 1967 (STI/PUB/143).

41. M. A. Bredig, J. W. Johnson, and W. T. Smith, Jr., "Miscibility of Liquid Metals with Salts. I. The Sodium-Sodium Halide Systems," J. Amer. Chem. Soc, **77**, 307-312 (January 20, 1955).

42. R. E. Mueller, R. J. Beaudry, and J. G. Feldes, "Results of Fission-Product Activity Analysis to Determine Extent of Fuel Failure in Fermi Accident," Trans. Amer. Nucl. Soc., **10**(2), 334-335 (November 1967).

43. R. R. Smith, C. B. Doe, and F. S. Kirn, "The Application of Cover-Gas Monitoring in the Recent Fission Product Release," Trans. Amer. Nucl. Soc., **10**(2), 634 (November 1967).

44. G. Batté and G. L. Hofman, "Run-Beyond-Cladding Breach (RBCB) Test Results for the Integral Fast Reactor (IFR) Metallic Fuels Program," International Fast Reactor Safety Meeting IV, p. 207, Snowbird, Utah, 12-16 August (1990).

45. J. Findlay, "The Migration of Fission Products through Reactor Fuel Materials," Proceedings of a Panel on the Behaviour and Chemical State of Irradiated Ceramic Fuels, Held in Vienna, 7-11 August 1972, IAEA-PL-463/10, pp. 211-220 (1974).

46. International Atomic Energy Agency, *Control of Iodine in the Nuclear Industry*, Report of a Panel on the Control of Iodine and Other Constituents of Airborne Radioactive Wastes Held in Vienna by the Atomic Energy Agency, Technical Report Series No.148. (1973).

47. M. Aubert, D. Calais, and R. Le Beouze, "Role de L'Iodde Sure Le Developpement De La Reaction Entre Le Combustible Et Sa Gaine In Acier Inoxydable (type 316) Das Les Reacteurs," J. Nucl. Materials, 58, 257-277 (1975).

48. K. K. Brown, Jr., "Re-examining the Significance of Fission Products in Accident Situations," Nucleonics, **23**, 55-59 (1965).

49. D. A. Collins et al. (UK AEA, Risley, England). Sampling and characterization techniques used in the study of iodine release from irradiated fuel elements. (1965). Citation in Reference 48.

50. R. R. Smith, D. W. Cissel, C. B. Doe, E. R. Ebersole, and F. S. Kirn, "Locating and Identifying the Source of the May 24, 1967 Fission-Product Release in EBR-II," ANL-7543 (April 1969).

51. R. S. Hart, "Fission Product Solubility in Sodium," in Annual Technical Progress Report, AEC Unclassified Programs, Fiscal Year 1961, NAA-SR-6370 (August 15, 1961), pp I-E-7 to I-E-9. BNA07659438.

52. R. S. Hart and C. T. Nelson, "Fission Product Retention Characteristics of Sodium at High Temperatures," NAA-SR-MEMO-8712 SPECIAL (February 1965). BNA03024721

53. W. P. Kunkel, A. S. Gibson, C. F. R. Smith, R. G. Begley, and L. Baurmash, "Fission Product Retention by Reactor Coolants Initial Report, NAA-SR-MEMO-7663 (undated; ≤1963). BNA8370457.

54. J. D. Christian and W. P. Gilbreath, "Defect Structure of NiO and Rates and Mechanisms of Formation from Atomic Oxygen and Nickel," Oxid. of Metals, 9, 1-25 (1975).

55. W. P. Kunkel, "Fission Product Retention in Sodium – A Summary of Analytical and Experimental Studies at Atomics International," NAA-SR-11766 August 8, 1966). BNA08443993.

56. "Quarterly Technical Progress Report, AEC Unclassified Programs July-September 1965," NAA-SR-11650 (December 1965), pp. V-7 to V-13. BNA07813714.

57. J. Saroul, "Investigation on the Behaviour of Fission Products in Sodium and Argon – PIRANA Experiments," Proc. International Conference on the Safety of Fast Reactors, Aix-en-Provence, September 19-22, 1967. pp. Vb-1-1 to Vb-1-10.

58. Figure A-IV-3, Gas Services, P and I Diagram (Sheet 2 of 3; 9693-70019), June 27, 1960, BNA00458114.

59. J. A. Daniel, Sr., "Investigation of Releases from Santa Susana Sodium Reactor Experiment in July 1959," prepared for Paul, Hastings, Janofsky & Walker, LLP, San Francisco, May 2005.

60. G. Borg, Interoffice letter to W. L. Fisher, "Quarterly report of activity released to atmosphere." November 20, 1959. BNA00485201.

61. J. Daniel, Daniel & Associates, personal communication to J. D. Christian.

62. E. E. Shpilrain et al., "Solubility of Noble Gases in Liquid Alkali Metals," paper presented at the Fourteenth Symposium on Thermophysical Properties, June 25-30, 2000, Boulder, Colorado. See http://www.symp14.nist.gov/PDF/MET02SHP.PDF.

63. E. Veleckis, F. A. Cafasso, and H. M. Feder, "Solubility of Xenon in Liquid Sodium," J. Chemical and Engineering Data, **21**, 75-76 (1976).

64. R. A. Blomquist, F. A. Cafasso and H. M. Feder, "Solubility of Krypton in Liquid Sodium," J. Nuclear Materials, **59**, 199-200 (1976).

65. "Plaintiffs' FRCP Rule 26(a)(2) Disclosure of Expert Testimony and Report of Arjun Makhijani," U. S. District Court, Central District of California, Western Division, Case No. 97-1554 DT (RCx), Lawrence O'Conner et al., Plaintiffs, v. Boeing North American, Inc., et al., Defendants (January 18, 2005).

66. R. S. Hart and H. H. Cappel, "Principal Uncertainties in Sodium-Graphite Reactor Systems Hazards Evaluations," NAA-SR-TDR-5643 (>May 1960). BNA02336583.

67. R. S. Neymark, "Rate of Alloying of Uranium Alloys with Stainless Steel /Part I. 1800 to 2300 Degree F." NAA-SR-3278. BNA0831490.

68. N. K. Simon, "SRE-PEP Fuel Handling Accident," North American Aviation, Inc. Internal Letter 2-241-64 to C. W. Griffin, dated January 11, 1964. BNA02008156.

69. Drawing A-IV-I, Primary-Sodium System, P and I Diagram (9693-70016), June 27, 1960, BNA0458111; and Drawing A-IV-3, Gas Services, P and I Diagram (Sheet 2 of 3; 9693-70019), June 27, 1960, BNA00458114. Drawings show cover gas line from reactor either entering top of primary sodium fill tank and exiting through the sodium vapor trap to the gas service system, or diverting through a line bypassing the sodium fill tank and passing through a second sodium vapor trap. These are the only routes for the cover gas.

70. G. E. Deegan et al., "Design Modifications to the SRE during FY 1960," NAA-SR-5358 (rev), June 1960. BNA07661532.

71. D. H. Johnson, "Pre-Operational Acceptance Test Procedures for the Sodium Reactor Experiment Prepared by Sodium Graphite Reactors Group," NAA-SR-MEMO-1681 (July 17, 1956. BNA03968609. See pp. BNA03968825 and BNA03968826.

72. J. O. Nicholson, "Sodium Reactor Experiment Operator Training Manual," NAA-SR-6895 (January 1965). BNA01134270.

73. U. S. Environmental Protection Agency web site: http://www.epa.gov/radiation/radionuclides/iodine.htm

74. ANSI N13.1-1969, "American National Standard Guide to Sampling Airborne Radioactive Materials in Nuclear Facilities."

75. J. D. McCormack, "I-131 Sampling System Efficiencies at PUREX and REDOX Stacks," HW-73288 (June 1, 1962).

76. ANSI/HPS N13.1-1999, "Sampling and Monitoring Releases of Airborne Radioactive Substances from the Stacks of Nuclear Facilities," Approved 12 January 1999, American National Standards Institute.

77. A. Makhijani, "Declaration of Arjun Makhijani, Ph.D.," U. S. District Court, Central District of California, Western Division, Case No. 97-1554 DT (RCx), Lawrence O'Conner et al., Plaintiffs, v. Boeing North American, Inc., et al., Defendants (February 11, 2004).

78. "Sodium Reactor Experiment Power Expansion Program. Reactor Safety Analysis Report, Draft" NAA-SR-9516 (May 9, 1964). BNA00456687.

79. "Sodium Reactor Experiment Power Expansion Program. Reactor Safety Analysis Report," NAA-SR-9516 (Rev.) (November 2, 1964). BNA08427366.

80. W. P. Kunkel, A. S. Gibson, , C. R. F. Smith, and R. J. Hegley, "Fission Product Retention by Reactor Coolants – Initial Report," NAA-SR-MEMO-7663 (no date; ≤1963). BNA08370457.

81. W. P. Kunkel, D. M. Elliott, and A. S. Gibson, "Fission Product Retention in Reactor Coolants – Results of the Second in-Pile Experiment," NAA-SR-MEMO-8247 (1963). BNA0837290.

82. W. P. Kunkel, D. M. Elliott, and A. S. Gibson, "Fission Product Retention in Sodium at 1000°F – The Third In-Pile Experiment," NAA-SR-MEMO-8801 (1963). BNA08374201.

83. Videotaped Deposition of Arjun J. Makhijani, Wednesday, March 9, 2005, Los Angeles, California, Volume 2, Reported by Darlene A. Matura, CSR No. 5959, Job No. 60938/908963.

84. L. L. Burger, "Fission Product Iodine During Early Hanford-Site Operations: Its Production and Behavior During Fuel Processing, Off-Gas Treatment and Release to the Atmosphere," PNL-7210 HEDR (May 1991).

85. D. D. Calkins, Interoffice Correspondence to B. R. Hayward, "SRE Fuel Committee Data," August 27, 1959. BNA02010126.

86. J. L. Ballif, "Interim Report on Fuel Damaged at the SRE during Run No. 14," NAA-SR-TDR-4431. BNA08449977.

87. G. Borg, “Wash Cell Incident at the Sodium Reactor,: AI-MEMO-5155 (April 6,1960). BNA07198106.

88. N. K. Simon, NAA internal letter to C. W. Griffin, "SRE-PEP Fuel Handling Accident," January 22, 1964. BNA02008156.

89. J. P. Hale, Atomics International internal letter to C. A. Willis, "Answer to DRL Safety Analysis Question 1 Core Meltdown Accident SRE-PEP," April 22, 1966. BNA02012265.

90. C. M. Heeb, "Iodine-131 Releases from the Hanford Site, 1044 through 1947, Volume 1 – Text," PNWD-2033 HEDR Vol. 1 (March 1993).

91. D. A. Miller and M. M. Smith, letter to J. M. Tilley, "Production of I^{131}," HW-7-4073 (April 17, 1946).

92. J. B. Work, letter to A. B. Greninger, "Disposal of Separation Plant Gases," HW-7-5520 (December 18, 1946). Work references J. H. Tilley to A. V. Peterson, May 21, 1946, File No. 8080.

93. R. S. Apple, Memorandum to File, "Recovery of I^{131}," HW-7-4074 (May 16, 1946).

94. J. L. Dreher, "The Evolution of Iodine During Metal Dissolution," Memorandum Report – SE-PC-#74, HW-3-3003 (August 2, 1945).

95. Exhibit 84 of Videotaped Deposition of Arjun J. Makhijani, Wednesday, March 9, 2005, Los Angeles, California, Volume 2, Reported by Darlene A. Matura, CSR No. 5959, Job No. 60938/908963. O. Schumacher, "Review of documents of the SRE incident on July 1959, assessment of possible source term to the environment," January 30, 2004.

A

APPENDIX A

EVALUATION OF SODIUM AND COVER GAS SAMPLE DATA – CORRELATIONS WITH HYPOTHETICAL RELEASE DATES OF FISSION PRODUCTS INTO THEM

Following the Run 14 incident, the sodium was first sampled on August 2, 1959, about seven days after reactor shutdown, and analyzed for major fission products. It was again sampled on October 31. When decay was taken into account, the amounts of the fission products present on October 31 were much smaller than calculated from the August 2 sample. The two exceptions were cesium and iodine, which were substantially still in solution.[A1, Tables IV and V] A summary of these results, taken from Hart's Table V, is given in Table A1. The August 2 sample results are expressed for what they would have been on July 26.

Table A1. SRE primary sodium August 2 sample results for fission products.

Isotope	Fission Product Contamination, µCi/g Sodium			Ratio $\frac{\text{Oct 31 (actual)}}{\text{Oct 31 (predicted)}}$
	Aug 2*	Oct 31 (predicted)	Oct 31 (actual)	
Cs-137	1.26	1.26	0.45	0.36
Cs-134	0.02	0.02	0.006	0.3
Sr-90	0.97	0.97	0.060	0.062
Sr-89	19.0	5.8	0.25	0.043
1-131	0.42	0.00019	0.00012	0.63
Ce-141	3.7	0.54	0.000088	0.00016
Ce-144	5.1	3.9	0.00031	0.00008
Zr-95+Nb-95	13.0	5.2	0.0067	0.0013
Ru-103	0.85	0.19	0.0045	0.024

*Calculated as of July 26.

During the almost three month interval, the cerium and zirconium/niobium isotopes have almost completely plated out in the system. Only a few percent of the strontium and ruthenium remain in solution or suspension.

If the fission products were released primarily from failures on July 23, which is ten days before the first sample, likely most of the cesium and iodine were still in solution, and a large fraction of the other isotopes may have been, as well. If the major release was on July 13, twenty days before sampling, a bit of the cesium may have been removed, more so than iodine, but the remaining quantities (corrected for decay) would likely be still fairly close to the original values on the date of release. Thus, one may derive percent releases of iodine and cesium from the failed fuels for the two postulated dates of release, using the sodium data of August 2.

The results of such a calculation, using inventories in the failed fuels calculated by Daniel & Associates[A2], are summarized in Table A2. The strontium results indicate that its plateout may not have been much more than cesium before August 2. The variability between Sr-89 and Sr-90 indicates the apparent precision of the measurements.

Table A2. Percent of isotopes from failed fuels in sodium calculated for July 13 and July 23 postulated release dates.

Isotope	Ci in Na*	July 13 Postulated Release		July 23 Postulated Release	
		Ci in Fuels**	% Release	Ci in Fuels**	% Release
Cs-137	27.7	2,526	1.10	2,555	1.08
Sr-89	444	39,410	1.13	41,710	1.06
Sr-90	21.4	2,511	0.85	2,526	0.85
I-131	16.3	1,254	1.30	5,905	0.28
I-133		0.561	1.30	2,023	0.28

*August 2 sample calculated for July 26.
**Inventories for the date decayed to July 26 to put on common basis with reported sodium data.

The cover gas sample taken on August 12 and analyzed on September 14 by gamma spectroscopy yielded the result reported by Hart[A1, p. 13] of 7.4 µCi/cm3 Xe-133 and 0.016 µCi/cm^3 Kr-26 calculated back to the shutdown date of July 26, which would be decayed to 0.010 µCi/cm^3 Xe-133 and 0.016 µCi/cm^3 Kr-85 on September 14 and to 0.78 µCi/cm^3 Xe-133 and 0.016 µCi/cm^3 Kr-85 on August 12, the date of sampling. (The interim report reports the September 14 measurements directly as 0.010 µCi/cm^3 Xe-133 and 0.016 µCi/cm^3 Kr-85.[A3, p. IV-C-15]) The August 14 total activity of 0.80 µCi/cm^3 determined from the gamma spectroscopy compares to 0.87 µCi/cm^3 gross activity reported for that date in the interim report.[A3, p. IV-C-14] This could be an indication that the Kr-85 counting by gamma spectroscopy could be somewhat low. Daniel has had extensive experience in gamma counting samples of Kr-85 and indicates that errors of this magnitude are not uncommon. The errors are always on the low side for Kr-85 amount.[4 The significance of this will be seen in the following analysis.

The inventories of Kr-85, Xe-133, and Xe-133m (which decays to Xe-133 with half life of 2.19 days) in the SRE failed fuel elements on postulated release dates of July 13 and July 23 (calculated by Daniel & Associates) are shown in Table A3. The I-133 in sodium, calculated as the percent of failed fuel inventory of I-131 found in sodium, Table A2, times the fuel I-133 inventory is also shown because it decays to Xe-133 with a half life of 20.8 hours.

Table A3. Fission products in failed fuels on postulated release dates of July 13 and July 23 potentially contributing to noble gases in cover gas, curies.

Isotope	Postulated Release Date	
	July 13	July23
Kr-85	316.8	318.1
Xe-133	8,779	37,380
Xe-133m	357.6	1,261
I-133*	239.0**	60.1*

*Decays to Xe-133.
**Fuel inventory of I-133 multiplied by the fractional release of I-131 to sodium, Table A2.

These results are decayed to September 14 and divided by the core cover gas volume as derived from the data of Hart (see Appendix A) to obtain their individual contributions to the cover gas sample and compared with the cover gas concentrations in Table A4. Of the original fuel Kr-85 and Xe-133, the fractions released to the cover gas are indicated as *y* and *z*, respectively.

Table A4. Xe-133 and Kr-85 concentrations in cover gas from failed fuels for postulated release dates of July 13 and July 23. Curie inventories in fuels decayed to September 14.

Isotope	Cover Gas, µCi/cm^3	From Fuels, µCi/cm^3	
		July 13 Release Date	July 23 Release Date
Kr-85	0.016	49.9 × *y*	50.0 × *y*
Xe-133	0.010	0.350 × *z*, from Xe-133	5.39 × *z*, from Xe-133
		0.0102 × *z*, from Xe-133m	0.130 × *z*, from Xe-133m
		0.00194, from I-133	0.00175
		0.360 × *z* + 0.00194, total Xe-133	5.52 × *z* + 0.00175, total Xe-133

By equating the total Xe-133 and the Kr-85 concentration expressions to the cover gas concentrations, *x* and *y* may be solved. Corrections to the results must be made for the fraction of cover gas estimated to have been vented prior to the sampling on August 12. From Table B1 in Appendix B, the most probable amount from July 13 to August 12 is 48% vented, 52% remaining. From July 23 to August 12, the probable venting is 16%, 84% remaining. Thus, the July 13 calculated *y* and *z* values are divided by 0.52 and the July 23 values are divided by 0.84. The results are summarized in Table A5 along with those from Table A2 for Cs-137 and I-131.

Table A5. Calculated percent releases of fission products from failed fuels for postulated release dates of July 13 and July 23.

Isotope	Calculated Percent Released from Failed Fuels	
	July 13 Release Date	July 23 Release Date
Xe-133	4.3	0.18
Kr-85	0.062	0.038
I-131	1.3	0.28
Cs-37	1.1	1.1

The I-133 contribution to the Xe-133 in the September 14 measurement is 19% for a July 13 release and 18% for a July 23 release.

This shows that the ratio of calculated Xe and Kr release fractions is 70 for an assumed July 13 release and 4.7 for an assumed July 23 release. As noted above, the Kr-85 cover gas sample results may be somewhat low, which if accommodated, would tend to bring the July 23 ratio closer to the expected unity.

APPENDIX A REFERENCES

A1. R. S. Hart, "Distribution of Fission Product Contamination in the SRE," NAA-SR-6890 (March 1962). BNA08401400.

A2. Daniel & Associates, Inc., 136 Parker Road, Crescent City, Florida 32112-4736.

A3. A. A. Jarrett, General Editor; R. L. Ashley et al., "SRE Fuel Element Damage, an Interim Report," NAS-SR-4488 (November 15, 1959). BNA03439250.

A4. J. A. Daniel, Daniel & Associates, Crescent City, Florida, personal communication to J. D. Christian.

APPENDIX A

EVALUATION OF SODIUM AND COVER GAS SAMPLE DATA – CORRELATIONS WITH HYPOTHETICAL RELEASE DATES OF FISSION PRODUCTS INTO THEM

Following the Run 14 incident, the sodium was first sampled on August 2, 1959, about seven days after reactor shutdown, and analyzed for major fission products. It was again sampled on October 31. When decay was taken into account, the amounts of the fission products present on October 31 were much smaller than calculated from the August 2 sample. The two exceptions were cesium and iodine, which were substantially still in solution.[A1, Tables IV and V] A summary of these results, taken from Hart's Table V, is given in Table A1. The August 2 sample results are expressed for what they would have been on July 26.

Table A1. SRE primary sodium August 2 sample results for fission products.

Isotope	Fission Product Contamination, μCi/g Sodium			Ratio
	Aug 2*	Oct 31 (predicted)	Oct 31 (actual)	Oct 31 (actual) / Oct 31 (predicted)
Cs-137	1.26	1.26	0.45	0.36
Cs-134	0.02	0.02	0.006	0.3
Sr-90	0.97	0.97	0.060	0.062
Sr-89	19.0	5.8	0.25	0.043
I-131	0.42	0.00019	0.00012	0.63
Ce-141	3.7	0.54	0.000088	0.00016
Ce-144	5.1	3.9	0.00031	0.00008
Zr-95+Nb-95	13.0	5.2	0.0067	0.0013
Ru-103	0.85	0.19	0.0045	0.024

*Calculated as of July 26.

During the almost three month interval, the cerium and zirconium/niobium isotopes have almost completely plated out in the system. Only a few percent of the strontium and ruthenium remain in solution or suspension.

If the fission products were released primarily from failures on July 23, which is ten days before the first sample, likely most of the cesium and iodine were still in solution, and a large fraction of the other isotopes may have been, as well. If the major release was on July 13, twenty days before sampling, a bit of the cesium may have been removed, more so than iodine, but the remaining quantities (corrected for decay) would likely be still fairly close to the original values on the date of release. Thus, one may derive percent releases of iodine and cesium from the failed fuels for the two postulated dates of release, using the sodium data of August 2.

The results of such a calculation, using inventories in the failed fuels calculated by Daniel & Associates[A2], are summarized in Table A2. The strontium results indicate that its plateout may not have been much more than cesium before August 2. The variability between Sr-89 and Sr-90 indicates the apparent precision of the measurements.

Table A2. Percent of isotopes from failed fuels in sodium calculated for July 13 and July 23 postulated release dates.

Isotope	Ci in Na*	July 13 Postulated Release		July 23 Postulated Release	
		Ci in Fuels**	% Release	Ci in Fuels**	% Release
Cs-137	27.7	2,526	1.10	2,555	1.08
Sr-89	444	39,410	1.13	41,710	1.06
Sr-90	21.4	2,511	0.85	2,526	0.85
I-131	16.3	1,254	1.30	5,905	0.28
I-133		0.561	1.30	2,023	0.28

*August 2 sample calculated for July 26.
**Inventories for the date decayed to July 26 to put on common basis with reported sodium data.

The cover gas sample taken on August 12 and analyzed on September 14 by gamma spectroscopy yielded the result reported by Hart[A1, p. 13] of 7.4 µCi/cm3 Xe-133 and 0.016 µCi/cm^3 Kr-26 calculated back to the shutdown date of July 26, which would be decayed to 0.010 µCi/cm^3 Xe-133 and 0.016 µCi/cm^3 Kr-85 on September 14 and to 0.78 µCi/cm^3 Xe-133 and 0.016 µCi/cm^3 Kr-85 on August 12, the date of sampling. (The interim report reports the September 14 measurements directly as 0.010 µCi/cm^3 Xe-133 and 0.016 µCi/cm^3 Kr-85.[A3, p. IV-C-15]) The August 14 total activity of 0.80 µCi/cm^3 determined from the gamma spectroscopy compares to 0.87 µCi/cm^3 gross activity reported for that date in the interim report.[A3, p. IV-C-14] This could be an indication that the Kr-85 counting by gamma spectroscopy could be somewhat low. Daniel has had extensive experience in gamma counting samples of Kr-85 and indicates that errors of this magnitude are not uncommon. The errors are always on the low side for Kr-85 amount.[4 The significance of this will be seen in the following analysis.

The inventories of Kr-85, Xe-133, and Xe-133m (which decays to Xe-133 with half life of 2.19 days) in the SRE failed fuel elements on postulated release dates of July 13 and July 23 (calculated by Daniel & Associates) are shown in Table A3. The I-133 in sodium, calculated as the percent of failed fuel inventory of I-131 found in sodium, Table A2, times the fuel I-133 inventory is also shown because it decays to Xe-133 with a half life of 20.8 hours.

Table A3. Fission products in failed fuels on postulated release dates of July 13 and July 23 potentially contributing to noble gases in cover gas, curies.

Isotope	Postulated Release Date	
	July 13	July23
Kr-85	316.8	318.1
Xe-133	8,779	37,380
Xe-133m	357.6	1,261
I-133*	239.0**	60.1*

*Decays to Xe-133.
**Fuel inventory of I-133 multiplied by the fractional release of I-131 to sodium, Table A2.

These results are decayed to September 14 and divided by the core cover gas volume as derived from the data of Hart (see Appendix A) to obtain their individual contributions to the cover gas sample and compared with the cover gas concentrations in Table A4. Of the original fuel Kr-85 and Xe-133, the fractions released to the cover gas are indicated as *y* and *z*, respectively.

Table A4. Xe-133 and Kr-85 concentrations in cover gas from failed fuels for postulated release dates of July 13 and July 23. Curie inventories in fuels decayed to September 14.

Isotope	Cover Gas, μCi/cm^3	From Fuels, μCi/cm^3	
		July 13 Release Date	July 23 Release Date
Kr-85	0.016	49.9 × *y*	50.0 × *y*
Xe-133	0.010	0.350 × *z*, from Xe-133	5.39 × *z*, from Xe-133
		0.0102 × *z*, from Xe-133m	0.130 × *z*, from Xe-133m
		0.00194, from I-133	0.00175
		0.360 × *z* + 0.00194, total Xe-133	5.52 × *z* + 0.00175, total Xe-133

By equating the total Xe-133 and the Kr-85 concentration expressions to the cover gas concentrations, *x* and *y* may be solved. Corrections to the results must be made for the fraction of cover gas estimated to have been vented prior to the sampling on August 12. From Table B1 in Appendix B, the most probable amount from July 13 to August 12 is 48% vented, 52% remaining. From July 23 to August 12, the probable venting is 16%, 84% remaining. Thus, the July 13 calculated *y* and *z* values are divided by 0.52 and the July 23 values are divided by 0.84. The results are summarized in Table A5 along with those from Table A2 for Cs-137 and I-131.

Table A5. Calculated percent releases of fission products from failed fuels for postulated release dates of July 13 and July 23.

Isotope	Calculated Percent Released from Failed Fuels	
	July 13 Release Date	July 23 Release Date
Xe-133	4.3	0.18
Kr-85	0.062	0.038
I-131	1.3	0.28
Cs-37	1.1	1.1

The I-133 contribution to the Xe-133 in the September 14 measurement is 19% for a July 13 release and 18% for a July 23 release.

This shows that the ratio of calculated Xe and Kr release fractions is 70 for an assumed July 13 release and 4.7 for an assumed July 23 release. As noted above, the Kr-85 cover gas sample results may be somewhat low, which if accommodated, would tend to bring the July 23 ratio closer to the expected unity.

APPENDIX A REFERENCES

A1. R. S. Hart, "Distribution of Fission Product Contamination in the SRE," NAA-SR-6890 (March 1962). BNA08401400.

A2. Daniel & Associates, Inc., 136 Parker Road, Crescent City, Florida 32112-4736.

A3. A. A. Jarrett, General Editor; R. L. Ashley et al., "SRE Fuel Element Damage, an Interim Report," NAS-SR-4488 (November 15, 1959). BNA03439250.

A4. J. A. Daniel, Daniel & Associates, Crescent City, Florida, personal communication to J. D. Christian.

B

APPENDIX B

COVER GAS VENTING AND FLUSHING EVALUATION AND ASSESSMENT OF ABILITY TO MEASURE I-131 IN COVER GAS SAMPLE

Venting Operations

In order to evaluate the venting and sampling data, the cover gas volume is required. The cover gas system consisted of the region above the reactor sodium pool, which was connected via a 50-foot 2-inch diameter pipe (31 L) to the sodium fill tank of volume 33,470 L[B1, Figure A-IV-1, p. BNA00458111] During normal operations after the sodium system had been filled, the fill tank was nearly empty, the sodium level being only 1 or 2 inches above the nozzle that fills the tank, so the gas volume is near 33, 300 L.

Sampling points were available to sample the core cover region where released radionuclides would be contained prior to any venting through the pipe to the fill tank and beyond. Hart[B2, p. 5] states, "Direct samples from the primary pool blanket gas may be taken by drawing the helium in an evacuated container from a suitable connection, usually the helium fill line." A valve between the core region and the fill tank would have been closed for sampling after shutdown of the reactor, see Figure C1, Appendix C. Thus, representative samples of the core region of the cover gas could be obtained.

The volume of the cover gas region above the core was somewhat variable during operations and was related to the height of sodium in the core. The nominal volume given in a safety evaluation of fission gas releases that may leak through the shield into the high bay was 9,150 L.[B3, p. B-1] Drawings on BNA pages 309 and 454 in the Operator Training Manual[B4] that show elevations and dimensions to the sodium surface indicate the design core cover gas volume to be 7,180 L. Based on data tabulated by Hart [B2,Table VII] for concentrations and total cover gas inventories of Xe-133 and Kr-85 gases sampled on August 12, 1959, it was either 6,300 L or 12,500 L on that date. Hart tabulated cover gas concentrations and total cover gas quantities for Xe-133 and Kr-85. The cover gas quantities included "a factor of two for probable losses due to pressure manipulations on July 18," which would indicate that he multiplied the calculated quantities using the actual cover volume by a factor of two. One calculates a volume of 6,300 L by dividing the Xe-133 quantity by its concentration and 12,500 L from the Kr-85 tabulated data. It would appear that Hart may not have multiplied the calculated Kr-85 quantity by two, which would favor the 6,300 L volume as being the correct value that he utilized. That is in line with the nominal design operating values listed above. We use that volume in our quantitative calculations, considering that it likely represented the sodium level throughout Run 14 at low power level.

The volume used by Hart is the core cover volume, without regard to the fill tank volume. Thus, for purposes of calculating Xe-133 and Kr-85 inventories in the cover gas, we shall accept Hart's 6,300 L volume, with any necessary corrections for venting operations prior to sampling. Any uncertainty in the volume used does not negate the general conclusions that can be drawn regarding whether or not the fission gases were completely removed from the cover gas by the venting/flushing operations and regarding the ability to measure I-131 in the sample when Xe-133 and Kr-85 were, in fact, measured.

The fraction of gas removed with each pressure release is calculated from the final (V_2) and initial (V_1) volumes by

$$\text{fraction removed} = \frac{V_2 - V_1}{V_1}$$

where the final volume is related to the initial volume by the two pressures before and after venting:

$$V_2 = \frac{p_1}{p_2} V_1$$

Thus,

$$\text{fraction removed} = \frac{p_1}{p_2} - 1$$

and fraction remaining = 1 – fraction removed = $2 - \frac{p_1}{p_2}$

The results of the following detailed discussions are summarized in Table B1, to aid in following them.

The cover gas was vented three times during the period July 12-15, referencing the Interim Report.[B5, pp. III-10 and III-13] At 15:30 on July 12, in an attempt to reduce the activity level in the reactor room/high bay area, the reactor pressure was lowered to less than 1 psig from its former pressure of 2 psig. Then, at 05:50 on July 15, in order to reduce the radioactivity level caused by xenon in the cover gas and determine the effect of pressure on core reactivity, the reactor atmosphere (cover gas) was reduced from 1.8 psig to 0.6 psig, repressurized to 3.0 psig, and then reduced to 1 psig. Converting each of these to absolute pressures by adding 14.7 psi, and conservatively assuming the first venting to 'less than 1 psig' was to 0.8 psig,* the calculated remaining fraction of original gas following the ventings is, using the above equations

$$0.923 \times 0.922 \times 0.873 = 0.742$$

Thus, these venting operations released only 26% of the original inventory present on July 12. Now, since the bulk of fission product release may have occurred on July 13 (following Makhijani's suggestion), the first factor does not apply to it and the net remaining inventory of it is 80%.

---

* Since the second venting specified the final pressure at 0.6 psig, if vented that low in the first venting, the value would likely have been specified. The selected 0.8 psig is half-way between 1 psig and 0.6 psig..

Table B-1. Summary of Cover Gas Venting Operations Relative to Postulated Core Activity Release Times and Sampling.

Date/Time	Operation	Fraction of Current Inventory Vented	Fraction of Current Inventory Remaining	Cumulative Fraction Vented (Cumulative Fraction Remaining)		Reason/Reference	Observations
				July 12 Release*	July 13 Release*		
July 12	Small leak to high bay					Interim Rept., p. III-10	At 15:30 high bay activity increased. At 16:20 air sampler filter showed 160,000 cpm. Hi radiation from core channel 7. Plugged channel 7 starting 2057.
July 12 after 1530	Vent 2 psig to ~0.8 psig	0.077	0.923	0.07 (0.923)	0 (1)	Reduce activity in high bay; Interim Rept, p. III-10	Sharp increase in stack activity at 17:00 to 1.5×10^{-4} µCi/cc; normal by 22:00. Due to direct radiation from high bay; ceased after plugged leak to high bay. Interim Rept.,p.IV-C-24
July 13 1825	Reactor excursion					Interim Rept., p. III-11	
July 14 0900	Small leak to high bay					Interim Rept., p. III-12	High bay activity increased to 14,000 cpm on air monitor. Localized to channels 29 & 50
July 15 0550	Vent 1.8 psig to 0.6 psig + pressurize to 3 psig, vent to 1 psig	0.196	0.804	0.258 (0.742)	0.196 (0.804)	Reduce xenon radioactivity in cover gas, improve core reactivity. Interim Rept., p. III-13	Starting at 06:00, stack monitor registered 7×10^{-5} Ci/cc intermittently until 1100. Interim Rept., p. IV-C-24
July 18	Sampled cover gas			0.258 (0.742)	0.196 (0.804)	Hart, p. 13. If sample was taken after the cover gas manipulation below, the remaining avg fraction would be 0.399	Hi radiation in sample. Estimated 2 to 10 µCi/cc, based on mostly Xe-133 ($t_{1/2}$=5.24 d) +Xe-135 activity ($t_{1/2}$=9.1 h).
				Wtd avg 0.202* (0.798)			
July 18	Manipulation of cover gas	0.50	0.50	0.629 (0.371)	0.598 (0.402)	Hart, p. 14, Table VII footnote	Hart applies this correction to the Aug 12 sample in interpreting results and does not mention the July 12,15, and 29 ventings.**
				Wtd avg 0.601 (0.399)			
July 19 & 20	Additional pressurization tests	$0^{\S}$ to 0.196	$1^{\S}$ to 0.804	$0.629^{\S}$ to 0.702 ($0.371^{\S}$ to 0.298)	$0.598^{\S}$ to 0.677 ($0.402^{\S}$ to 0.323)	To test effect on core reactivity. Interim Rept, p. III-14	***No mention of venting, just pressurization.*** Conservative calculations assume venting same as on July 15.

Date/Time	Operation	Fraction of Current Inventory Vented	Fraction of Current Inventory Remaining	Cumulative Fraction Vented (Cumulative Fraction Remaining)		Reason/Reference	Observations
				July 12 Release*	July 13 Release*		
July 26	Shut down reactor. Began bleeding & flushing to gas decay tanks almost immediately			0.629[§] to 0.702 (0.371[§] to 0.298)	0.598[§] to 0.677 (0.402[§] to 0.323)	Hart, p. 13.	Between 34 and 41% of the cover gas remained when shut down.*,** It is quite probable that the percent remaining was twice these numbers.
				Wtd avg 0.59[§] to 0.68 (0.41[§] to 0.32)			
July 29	Cover gas bleed down "to negative"	0.18	0.82	0.695[§] to 0.755 (0.305[§] to 0.245)	0.670[§] to 0.734 (0.330[§] to 0.266)	Interim Report., Table IV-C-8	Assumes an unspecified pressure change from 1.8 to -0.7 psig.
				Wtd avg 0.67[§] to 0.74 (0.33[§] to 0.26)			
August 12	Cover gas sampled			Wtd avg 0.67[§] to 0.74 (0.33[§] to 0.26)			***A minimum of between 26 and 33% of originally released fuel fission gases remain in sampled cover gas (before decay corrections). The more probable value is twice that.***

*July 12 release is considered relatively small. The July 13 release during the core excursion is considered the majority source of fission product release for the purpose of evaluating Makhijani's theory. Therefore, the weighted average for cumulative releases is based on the reasonable assumption that 10% of the releases occurred on July 12 and 90% on July 13. Then, a minimum of 26% of the cover gas fission gases would have remained in the core cover gas when it was sampled on August 12. A probable value (see next footnote) was 53 to 65%, depending on whether the July 19 and 20 pressure tests included venting.

**It is entirely possible that Hart misread an entry in Table IV-C-8, "Activity History of the Reactor Cover Gas," in the interim report on July 29 for July 18. The table states that the reactor cover gas was bled down to negative on July 29 and does not indicate any cover gas operations in the July 18 entry just above it, the date that Hart refers to. For conservativeness, we will include all mentioned operations to derive a minimum remaining fraction of cover gas when sampled on August 12, which turns out to be 26%. to report along with the probable fraction on August12 remained on September 29, i.e., 75% was purged out between the dates of the August 12 and September 29 samples.

[§] Based on not venting, just pressurization for tests on July 19 and 20.

The major release is considered by Makhijani to have occurred during the July 13 reactor excursion. A small amount may have occurred on July 12. It is likely that most of the thirteen fuel failures occurred in July 21-24 time frame.[B5, pp. IV-C-21 and B6] This is discussed in Section XIII of the text. As a practical conservative estimate for modeling purposes to evaluate Makhijani's concerns regarding the ability to measure fission gases in the cover gas sample from his postulated releases, we shall consider that 10% of the fission product release from failed fuels occurred on July 12 and 90% on July 13. This results in a maximized estimate of the quantities that would have been swept out of the cover gas before sampling to give Makhijani the maximum benefit of doubt. Using this weighted average vented fission gases from July 12 and 13 releases from the fuel would be 20.1% (79.9% retained) following the July 15 venting.

Hart applied a correction to the results of the August 12 sample, multiplying them by two "for probable losses due to pressure manipulations on July 18."[B7, p. 14] He did not mention in conjunction with this either the July 12 and 15 ventings or ventings subsequent to July 26. Thus, there is some confusion from the different statements. As a *worst case*, if one adds this 50% reduction on July 18 to the others calculated above, the remaining gases as of July 18 amount to 40% of the original inventories that would have been released from the failed fuels. However, the probable remaining amount is 80% – see second footnote of Table B1.

Then, on July 19 and 20, some additional tests were made in an attempt to evaluate the effect of core cover gas pressure on reactivity.[B5, p. II-14] No description of the procedure is given and the statement does not specifically say that any gas pressure relief was done. If one, as a conservative measure, assumes the same procedure as was done on July 15, the factor of 0.80 would be applied to fraction of gas retained following the operations. Thus, somewhere between 20 and 0% of the gas inventory remaining after the July 18 pressure manipulations (about 40% of the original at that time) was released on July 19 and 20, that is, between 80 and 100% of the July 18 inventory remained following July 20, or 30 to 40% of the original inventory from July 12 an 13.

A reasonable approximation of the released inventory fraction from the failed fuel elements that remained in the cover gas by July 26 is, then, about 30 to 40%. Hart states that bleeding and flushing operations to the gas decay tanks were almost immediately commenced following shutdown on July 26, but he does not give specific information. The quantitative flushings can be estimated from other data provided by Hart. A gamma scan analysis on September 14 of a cover gas sample taken on August 12 determined the presence of Xe-133 and Kr-85. Extrapolating the data back to the shutdown date of July 26 (i.e., correcting for decay) yielded 7.4 μCi/cc Xe-133 and 0.016 μCi/cc Kr-85. The Xe-133 concentration, while acknowledged to be somewhat lower than what corresponds to actual releases, is in rough agreement with the estimate made from a sample taken on July 18, which had an estimated content of 2 to 10 μCi/cc, assuming the principal activity to be Xe-133 (half life, 5.24 days) and Xe-135 (half life, 9.2 hours).[B7, p. 13] This indicates that not a substantial amount of flushing out occurred between July 26 (nor July 18) and August 12.

On July 29, a cover gas bleed down "to negative" was done.[B5, Table IV-C-8] The pressure changes were not specified. As a conservative basis, we assume 1.8 psig (16.5 psia) as the starting pressure, the same as on July 15. The final pressure is assumed to be slightly more than the reduced pressure of the pressure suction tank to which the gases are transferred, which operated at about -1.5 psig (13.2 psia). (Note that "negative pressure" denotes pressure relative to ambient pressure, i.e. gauge pressure. It means that the pressure in the vessel is less than the pressure outside. There is no such thing as a negative absolute pressure.) The actual pressure was probably barely negative gauge pressure, within -0.5 psig or so, with a slight overshoot of 0 gauge pressure as gas was transferred to the suction tank. A highly negative pressure would be avoided in order to prevent suction of air into the core cover region from the high bay. We conservatively assume -0.7 psig. From these, one calculates a release of 18% of current inventory and the remaining original gases to be 26 to 33% when this is applied to the July 26 inventory. This would be what was present as a minimum at the time of the August 12 sampling.

Given the comparable values of Xe-133 calculated from both the July 18 and August 12 samples, the remaining fractions calculated above of 0.40 on July 18 and 0.26 to 0.33 on August 12 are consistent.

A summary of these pre-August 12 cover gas manipulations that would affect the quantity of fission gases released to the cover gas being present when it was sampled on August 12 is provided in Table B-1. The data show that the fission gases were not quantitatively flushed out of the cover gas prior to obtaining the sample and that at least 26%, and more probably 52%, of the original xenon, krypton, and iodine (before decay corrections, addressed later) would have been in the sample were they released from the fuels on July 12 and 13.

The reasonableness of this estimate is supported by the fact that approximately ten subsequent flushings occurred over the next seven weeks, compared with the small amount of venting that occurred in the first 2½ weeks, calculated from quantitative data provided by Hart. "Through radioactive decay of the Xe-133 and cover gas purging operations, the fission product contamination level in the reactor cover gas was reduced to 4×10^{-3} μCi/cc (Kr-85) by September 29, 1959."[B7, p. 13] This is a reduction from 0.016 μCi/cc Kr-85 calculated for July 26, 1959 from a sample taken on August 12. Thus, $4\times10^{-3}/0.016 = 0.25$ fraction of the Kr-85 on August 12 remained on September 29, i.e., 75% was purged out between the dates of the August 12 and September 29 samples.

The operating procedures[B8, pp. V2-V3] specify the flushing procedure as follows. After opening the appropriate valves, the helium flow was adjusted to 1.5 scfm for approximately 20 min. This would purge 30 standard cubic feet, or 850 L of cover gas. The total cover gas volume above the core was between about 6,300L, so a single flush would (assuming plug flow that would result in a conservative maximum value) remove about 13.5% of the gas.[•] This corresponds to about 10 flushings for the 75% purging between August 12 and September 29 $[(1 - 0.13)^{10} = 0.25$ fraction remaining]. These additional extensive flushings indicates substantially incomplete flushing as of August 12.

[•] A more rigorous calculation based on the formula provided by R. L. Ashley et al., "Evaluation of the Atomics International Nuclear Development Field Laboratory as a Location for Reactor Facilities," R. L. Ashley, General Editor, NAA-SR-7300 Special (May 25, 1962), BNA08401054, p. 42 yields 12.6% removal for cover gas volume of 6,300 L. The formula is

$$C(t) = C_0 e^{-Rt/V}$$

where C_0 is the initial concentration and C(t) is the concentration after purging a volume, V, at a purge rate, R, for a time, t.

A calculation of the fraction of gas flushed out per purge can also be made from data reported in Table IV-C-8 of the interim report on August 12 and August 28. It is stated that the cover gas was purged on August 19 and 20, from which one calculates 23% flushed, using the purge procedure discussed earlier (13% removed per purge: 0.87×0.87 = 0.77 remaining). By comparing the ratios of activities reported on the two dates with the ratios calculated from the quantities of Xe-133 and Kr-85 measured in the August 12 sample and applying decay corrections, one derives that 29% of the gas was flushed between those dates in order to get down to the reported activity, very consistent with the calculation using the individual purge fractions. Thus, the purge calculations are validated.

By doing similar comparisons of the ratio of activities for the dates September 16, when cover gas purging was started, and September 29 reported in Table IV-C-8 with calculated activities from the August 12 sample corrected for decay, one calculates that five purges were done from September 16 to 20. Added to the two purges on August 19 and 20, the total is 7, which compares fairly well with the 10 estimated above* for between August 12 and September 29.

Therefore, the reasonable conclusion is that about 26 to 33% (and probably 52 to 66% if Hart erred in his statement of a factor of two flushing on July 18) of the original cover gas activity on July 12 and 13 remained when the sample was taken on August 12 (before decay corrections). That is why Xe-133 and Kr-85 were measured in the August 12 sample when it was gamma scanned on September 14. If the majority of the noble gases were released from the fuels on about July 23, as data indicate, then only 18% was vented prior to sampling.

Cover Gas Sample Analysis

What can be said, then, regarding the absence of detectable I-131 when the August 12 sample was gamma scanned on September 14? The September 14 scan showed the presence of 0.010 μCi/cc Xe-133 and 0.016 μCi/cc Kr-85.[B5, p. IV-C-15 These extrapolate back to 7.4 μCi/cc Xe-133 and 0.016 μCi/cc Kr-85 corrected for decay back to July 26[B7, p.13, B5, p. IV-C-15]. Now, on July 13, when most of the fuel failures are postulated by Makhijani to have occurred, the reactor inventory is representative of the relative quantities of the released gases that would be in the cover gas. These have been calculated by Daniel & Associates.[B9] On July 13 at 6:24 p.m. about when the power excursion occurred the core inventory of the thirteen failed elements was 3,845 Ci I-131, 18,390 Ci I-133, 8,779 Ci Xe-133, and 316.8 Ci Kr-85. I-133 is listed because the 20.8 hr half-life isotope decays to Xe-133 and must be accounted for in decay calculations. If one assumes, as does Makhijani, that the same fractions of xenon, krypton, and iodine were released from the fuels, which is 45% for his probable release, and that a net of 34.2% of the I-131 (his best estimate) was eventually released to and remained in the cover gas, the ratio of I-131-to-Xe-133 activities on July 13 would have been 0.334 and of I-131-to-Kr-85 activities would have been 9.22. Considering that venting and flushing activities would have purged out equal fractions of the gases, this ratio (before decay correction calculations) would

---

* Hart considered that the 0.004 μCi/cc remaining on September 29 was entirely due to Kr-85, whereas the detailed assessment here using the composition of the August 12 sample indicates half was due to Xe-133. If one applies that factor to the earlier calculation, the 10 flushings calculated from the Hart information become five, more in line with the estimate of this paragraph.

remain the same after those operations. Xe-133 has a half-life of 5.245 days and I-131 has a half-life of 8.04 days. Thus, Xe-133 decays away faster than does I-131. Taking into account the decays of the three isotopes I-131, I-133, and Xe-133 to September 14, one calculates the following activity (curie) ratios in the sample on September 14:

I-131/Xe-133	4.23
I-131/Kr-85	0.0408

Thus, since Xe-133 and Kr-85 were measured, if xenon, krypton, and iodine had been released and remained in the cover gas in the probable fractions claimed by Makhijani, the iodine would have been measured. It was looked for and not found, indicating its complete absence.

This consideration of measurement of I-131 relative to Xe-133 and Kr-85 may be quantified. By multiplying the activity concentrations of each isotope by its relative maximum gamma peak intensity per unit activity, one derives the following relative peak heights for the sample analysis on September 14 (accounting for peak overlaps and the relative concentrations of Xe-133 and I-131), based on Makhijani's postulated releases:

I-131 peak/Xe-133 peak	9.4
I-131 peak/Kr-85 peak	7.7

Even if only 3.6% (= 34.2%/9.4) of the I-131 had been released to and remained in the cover gas while 45% of Xe-133 and Kr-85 are assumed to have been released (though they were not, in fact, substantially released), its maximum peak height would have been the same as the Xe-133 peak height and would have been observed. Again, it must be emphasized that I-131 was looked for and not found. This is clear evidence that I-131 was NOT released to the cover gas.

The helium cover gas was periodically sampled during normal operations and assayed for radioactive content. Xenon-133 was always the dominant radioactive contaminant.[*] Attempts to detect iodine invariably gave negative results.[B7, p. 6] The typical Xe-133 concentration was on the order of 10^{-3} to 10^{-4} μCi/cc, compared with the 1.0×10^{-2} μCi/cc measured on September 14, a factor of up to 100 lower. This lends even more credence to the above calculation that the sensitivity of the measurements would have resulted in quantitative detection of any I-131 that would have been present on September 14. It means that I-131 present at 4.4%/100 = 0.04% of inventory in the failed elements would have been detected and measured on September 14. These observations during normal operations where I-131 was never seen in the cover gas are consistent with the September 14 sample analysis.

---

[*] The source was considered to be either pinhole leaks in the cladding or, more probably, from tramp uranium impurity in the sodium that produced fission products.

APPENDIX B REFERENCES

B1. R. E. Durand et al., "SRE Standard Operating Procedures," NAA-SR-MEMO-5326 (June 27, 1960). BNA00457851. Figure A-IV-1, Primary-Sodium System, P and I Diagram (9693-70016), June 27, 1960, p. BNA00458111.

B2. R. S. Hart, "Distribution of Fission Product Contamination in the SRE," NAA-SR-6890 (March 1962). BNA08401400.

B3. R. L. Ashley et al., "Evaluation of the Atomics International Nuclear Development Field Laboratory as a Location for Reactor Facilities," R. L. Ashley, General Editor, NAA-SR-7300 Special (May 25, 1962). BNA08401054.

B4. J. O. Nicholson et al., "Sodium Reactor Training Manual," NAA-SR-6895. BNA01134270.

B5. A. A. Jarrett, General Editor; R. L. Ashley et al., "SRE Fuel Element Damage, an Interim Report," NAS-SR-4488 (November 15, 1959). BNA03439250.

B6. F. L. Fillmore, "Analysis of SRE Power Excursion of July 13, 1959," NAA-SR-5898 (September 15, 1961). BNA00617492.

B7. R. S. Hart, "Distribution of Fission Product Contamination in the SRE," NAA-SR-6890 (March 1962). BNA08401400.

B8. R. R. Durand et al., "SRE Standard Operating Procedures," NAA-SR-MEMO-5326 (June 27, 1960). BNA00457851.

B9. Daniel & Associates, Inc., 136 Parker Road, Crescent City, Florida 32112-4736.

C

APPENDIX C

ESTIMATE OF QUANTITY OF IODINE-131 IN COVER GAS VENTED BY JULY 15 IF MAKHIJANI'S THEORY WERE CORRECT

The 6,300 L core cover gas was connected to the 33,300 L sodium fill tank by a 50 foot long 2-inch diameter pipe. A depiction of the system is given in Figure C1 with vessels approximately to scale. Valves V137 and V496 in lines B and C are always open during reactor operation.

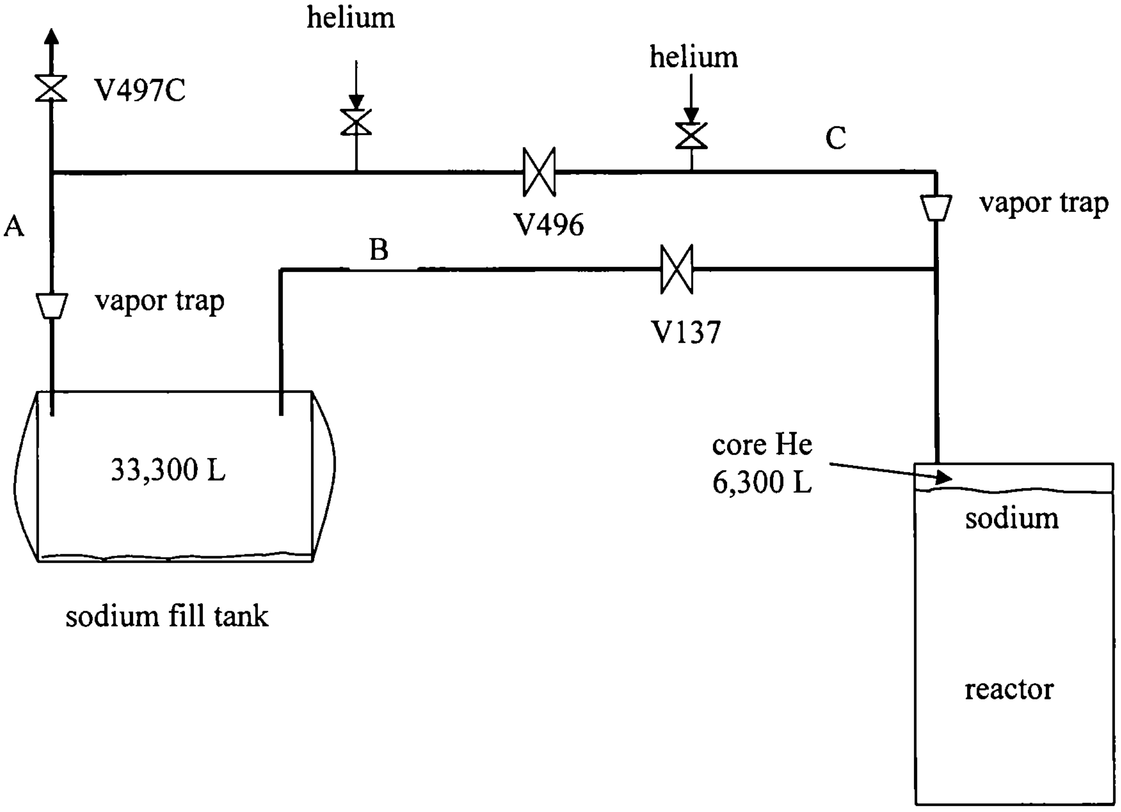


Figure C1. Cover gas system showing core and fill tank portions.

When venting after pressurization, valve V497C is opened. The fill tank can relieve pressure by transporting gas through A or through B and C. Since the gas volume there is much greater than in the core region, its mass flow will predominate. Any flow through B will simply slow down the venting of the core cover gas via C. Ultimately, the same percent of gas from each region will have been vented via V497C. Thus, the percent fission gases, which are in the core region only, vented is the same as the percent helium vented.

When the pressure is changed in the system, for example, reduced, the pressure above the core is relieved by venting some gas to the final pressure. The fraction of the volume transferred, ΔV/V, is calculated from

$$\frac{\Delta V}{V} = \frac{V_2 - V_1}{V_1} = \frac{\left(\frac{p_1}{p_2}\right)V_1 - V_1}{V_1} = \frac{p_1}{p_2} - 1$$

During the operations on July 12 and 15, the ratio of initial pressure, p_1, to final pressure, p_2, ranged from 1.08 to 1.13 and, therefore, between 8 and 13% of the volume of the core cover gas, that is, between 500 and 800 L was transferred out in any individual pressure relief. To calculate the fraction of fission gas in the core region that vented following pressurization, we assume, conservatively, that before the next pressure relief, the inventory in it has become uniformly mixed. This maximizes the quantity of iodine calculated to be vented from core region.

We start with Makhijani's assumption that the probable amount 1,330 Ci of I-131 was released from the failed fuel elements primarily before July 15 and resided in the core cover gas and insert the approximation, based on his considerations, that about 10% of that occurred on July 12 and 90% occurred during the July 13 excursion. Then the number of curies that would have been vented out of the core region (so as to be potentially available for release to the stack as asserted by Makhijani) is modeled for the three venting operations on July 12 and July 15, which Makhijhani claims resulted in complete release of the 1,330 curies.

The data are the following.

- July 12 15:30 133 Ci I-131 assumed released and remained in core cover gas until vented. Of that not vented on July 12, 20.3% decays by the July 15 venting.
- July 13 18:25 1,197 Ci I-131 assumed released and remained in core cover gas until vented. This decays to 1,057 Ci by the time of the July 15 venting.
- On July 12 at about 15:30, the pressure is changed from 16.7 psia to 15.5 psia.
- On July 15 at 05:50, the pressure is increased from 15.5 psia to 16.5 psia and then vented down to 15.3 psia.
- Again, on July 15, the pressure is increased from 15.3 psia back to 17.7 psia and then vented down to 15.7 psia.

These operations result in calculated transfers summarized in Table C1. A total of 236 Ci I-131 of Makhijani's postulated 1,330 Ci is estimated to have been vented from the core region and potentially released toward the stack, or 17.4% of the 1,330 Ci Makhijani postulated. (The final curie inventories tabulated for the core, fill tank, and vented out of the fill tank, as percent of original do not sum to 100% because of decay effects.)

Table C1. Iodine-131 that would have been vented from cover gas from July 12 and 15 venting operations.

Operation or occurrence	**Fraction of I-131 transferred out (or Ci I-131 released from fuel)**	**Ci transferred out of core gas**	**Ci remaining in core gas**
July 12 fuel release	133 Ci in fuel released to core cover		133 Ci
July 12 vent	0.0774	10.3 Ci	122.7 Ci; decays to 97.8 Ci July 15
July 13 excursion	1,197 Ci added to core cover; decays to 1,057 Ci July 15		1,155 Ci July 15
July 15 first venting	0.0784	90.5 Ci	1,064 Ci
July 15 second venting	0.127	135 Ci	929 Ci = 69.8% of original
Cumulative vented I-131		236 Ci = 17.4% of original	

These estimates are likely high because complete mixing of the fission gases with newly introduced helium may not have occurred before subsequent pressure relief. In addition to mixing time constraints, a factor is that an iodine molecule is 64 times heavier than a helium atom, which would tend to make the I_2 tend to remain toward the bottom of the core cover region. Therefore, it is possible that less than 236 Ci I-131 out of the postulated 1,330 Ci would have been vented out of the cover gas region. These calculations serve to establish a bracketing value based on the venting operations.

Comparable values of modeled I-131 releases to the stack based on Makhijani's postulated minimum and maximum bounding quantities in the cover gas (366 and 2,544 Ci, respectively,) are 65 and 450 Ci.

Note, also, that the path for iodine release is through a sodium vapor trap, which would tend to remove any iodine by reaction with sodium as well as deposition on metal surfaces.

It must be emphasized that this calculation of potential release of I-131 from the cover gas does not represent what actually happened. Based on the evaluations in the report, no I-131 is projected to have been released from the metal fuel, through the sodium, from the cover gas, through the sodium vapor trap, and to the stack without being substantially detected in the stack monitor and without being measured in the cover gas sample. The assessment here is of what the consequences of Makhijani's postulated inventory in the cover gas would be in terms of the venting activities.

D

APPENDIX D

IODINE-SODIUM SURFACE REACTION RATE EXPERIMENT

(J. R. Begley)[D1, pp. 18-22]

This experiment addressed the situation for a proposed sodium-cooled reactor with vented fuel elements in which fission gases would be vented through a tube from the fuel rods to the cover gas region in order to prevent pressure buildup in the rod plenum at high burnups. The consideration was for removal of iodine from the cover gas by reaction of the iodine gas with the sodium pool surface. Although the summary report by Kunkel of the previously unpublished results was published in 1966, Begley performed the experiments in 1964. From reports in 1962 by Castleman and in 1964 by Parker on characterization of iodine released from fuels under various conditions Begley recognized that iodine may exist in a number of complex chemical forms with uranium or other fission products. Nevertheless, the experiments were intended to address the possibility of elemental iodine gas being vented to the cover gas.

The experiment measured the rate of iodine loss from the vapor phase above a sodium surface at 500°F (260°C). The entire apparatus was maintained at isothermal conditions to reduce iodine condensation. Prior to heating, it was evacuated. When only I_2 was introduced above the sodium at a constant pressure of 2psia, the reaction rate was 4.5 mol/hr-ft^2. This would have included effects of any reaction with the sodium vapor, which was considered to be negligible. For reference, the quantity of I_2 corresponding to Makhijani's proposed release is 0.69 g or 0.0027 mol. The surface area of the SRE sodium pool was 95 ft^2. Thus, it is readily seen, in the absence of a limiting mass transfer rate, that the I_2 would have reacted almost instantly, a few thousandths of a second.

Begley then modeled the diffusion rate of I_2 in nitrogen, N_2, for which he had available diffusion coefficients (which were not available for I_2 in helium, He, and which would be greater). He determined that, for a cover gas depth of 2 feet (about the same as in the SRE) after a few hundredths of a second, the mass diffusion rate became much slower than the above reaction rate and was controlling. (His modeled system contained sufficient iodine to require more than that time for reaction directly with sodium.) The value of the average concentration as a function of time, relative to the initial concentration, of I_2 in the cover gas of 2-foot depth as a result of mass diffusion to and reaction with the sodium pool surface is given by

$$\frac{C_{avg}}{C_o} = 1 - 2(Dt)^{1/2}\left(\pi^{-1/2} + 2\sum_{n=1}^{\infty}(-1)^n ierf \frac{n}{(Dt)^{1/2}}\right)$$

where D = diffusion coefficient = 0.225 cm^2/sec for I_2 in N_2 at 500°F
C_o = initial concentration of I_2 in cover gas
C_{avg} = average concentration of I_2 at time t
t = time, sec

The ratio C_{avg}/C_o that represents the fraction of the original iodine remaining at time t is independent of the starting concentration in the system.

The results of the calculation as a function of time are shown in Figure D1. This fraction of initial iodine remaining is independent of initial concentration. The graph shows that after ½ hour, 50 percent of the iodine would diffuse to the surface and after about 3½ hours, 90 percent would. Virtually all would be gone in about 7¾ hours. In helium, the time would be substantially less because the diffusion coefficient would be greater.

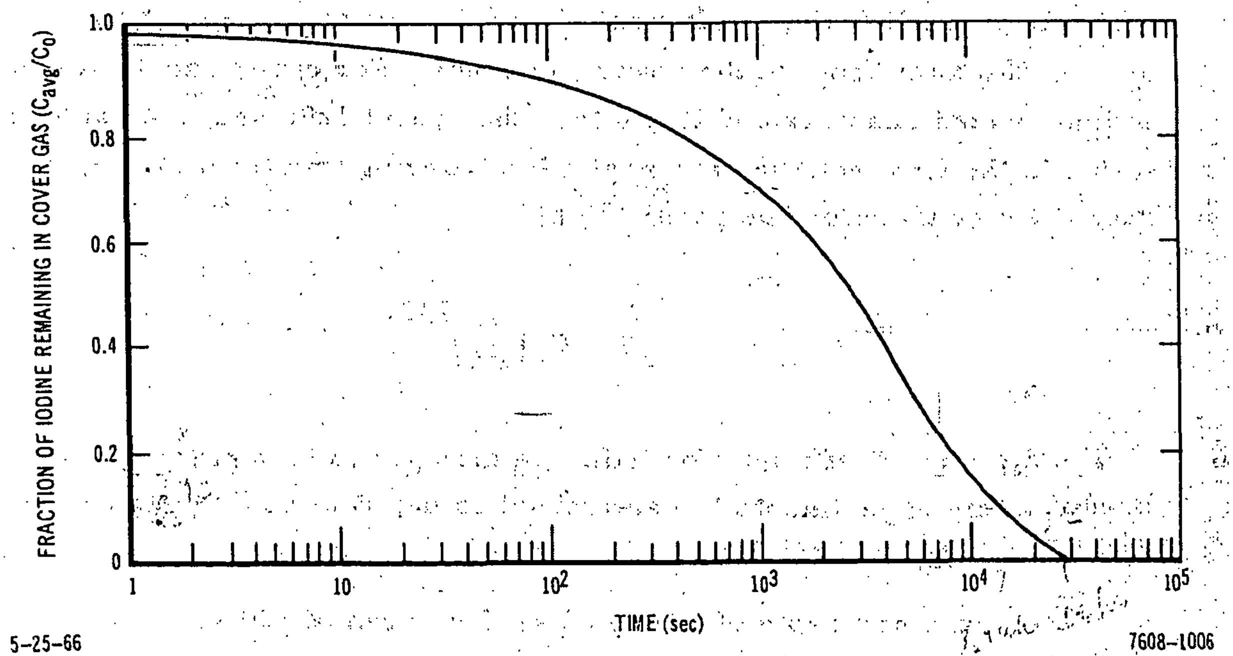


Figure 4. Rate of Iodine Diffusion to Sodium Surface

Figure D1. Removal rate of I_2 from a 2-foot depth of nitrogen above a sodium surface at 500°F.

Factors that would affect the rate in the SRE during the July 13 excursion in SRE are: 1) The sodium pool surface temperature was a minimum of 541°F. The temperature in the cover gas would range from that to somewhat less at the walls and top. The overall temperature effect would probably be relatively small. 2) The I_2 diffusion coefficient in helium would be more than twice that in nitrogen (approximately proportional to inverse of the square root of molecular weights, which is 2.65) 3) sodium condensed on the walls and ceiling of the core region above the sodium pool would decrease the effective distance by a factor of two for I_2 molecules to reach liquid sodium 3) Convection currents in the temperature gradient of the cover gas would add substantially to the mass transfer. 4) Reaction of I_2 with Na vapor atoms would add to the removal rate. While the gas phase reaction is much less than reaction rate at the pool surface, if may be significant relative to the diffusion rate to the surface. All these factors taken together would conservatively have resulted in removal times of one-fifth or less of those predicted by Figure B1: 6 minutes for 50% removal; 45 minutes for 90% removal; and 1½ hours for complete removal.

APPENDIX D REFERENCE

D1. W. P. Kunkel, "Fission Product Retention in Sodium – A Summary of Analytical and Experimental Studies at Atomics International," NAA-SR-11766 August 8, 1966). BNA08443993.

E

APPENDIX E

STACK MONITOR DETECTION OF FISSION GASES AT QUANTITIES SUGGESTED BY MAKHIJANI

Dr. Makhijani claims that I-131 and the noble gases Xe-133 and Kr-85 were passed by the stack monitor without detection in large part because of errors in attenuation factors used for the beta particles for the Geiger-Müller detector. The specific errors are provided in Section XXI.C of the report by analyzing the attenuation factors that he presented and relies on. The derivation of the relative contributions to the stack monitor detector of the gases that would have been present if Makhijani's theory were true is provided here. This is done by multiplying the number of curies of each contributing isotope by its attenuation factor for the detector. Those attenuation factors are given in the legend of Figure 13 of the report.

The source term of each contributing isotope for the 13 failed fuels is provided by Daniel & Associates for July 13 at the time of the excursion that Makhijiani purports to be the time of the majority of fuel failures and fission product releases.[E1] Those values are decayed to the time of the July 15 cover gas venting that Makhijani claims resulted in complete release of the cover gas to the stack. The fractions released from the core and retained in the cover gas until the venting as provided by Makhijani are used here to calculate the amount of the inventories supposedly vented: 45% of noble gases and 34.2% of iodine.[E2, p. 91] In addition to I-131, the contributions of I-133, I-135, and Xe-135 are included, since they would have followed the paths of I-131 and Xe-133. I-131 also decays to Xe-133, so adds to its quantity on July 15.

The complete compilation of the relevant data is summarized in Table E-1, in which the relative contributions to the Geiger-Müller detector are derived and tabulated. The relative signal for each isotope is its inventory divided by the attenuation factor.

These show that iodine would have contributed 51% of the signal, primarily because of the presence of I-133, if Makhijani's theoretical releases had occurred.

The total apparent overall attenuation factor is 9,961/1,424 = 7.00. The attenuation factor applied to the measurement was 5.59. That is, when calculating the inventory from the signal, its calibrated value was divided by 5.59 instead of the true 7.00 that would have been correct for this hypothetical mix of isotopes. This means that the calculated inventory would have been only 5.59/7.00 = 80% of the hypothetical inventory. That is, the reported inventory would be 20% low. Makhijani contends (without evaluating in a quantitative manner the measurement biases that he raises issue with) it was 99.9% low, an assertion that cannot be defended in the face of the quantitative analysis of the stack monitoring system.

Table E1. Stack monitor Geiger-Müller detector relative signals of isotopes in Makhijani's postulated vented fission gases on July 15.

Isotope	Failed fuel inventory during July 13 excursion, Ci	Cover gas inventory, July 13, Ci*	Cover gas inventory when vented July 15	Geiger-Müller detector attenuation factor	Relative G-M signal from the isotope
Kr-85	316.8	142.6	142.5	17	8.38
I-131	3,845	1,315	1,156	10	116
I-133	18,390	6,289	1,904 I-133 631.0 Xe-133	3.77 I-133	505 I-133
I-135	29,861	10,212	237.9 I-135 1,140 Xe-135	2.3	103 I-135
Xe-133	8,779	3,950	3,242		
Xe-133m	357.6	160.9	22.8 Xe-133		
Total Xe-133		3,950	3,896	14	278.3
Xe-135	50,081	22,536	1,473		
Xe-135m	13,230	5,954	11.3 Xe-135		
Total Xe-135			2,624	6.35	413
TOTAL			9,961		1,424

*Based on 45% of noble gases Kr and Xe and 34.2% of iodine isotopes from the failed elements being released to the cover gas and retained there until venting, per Makhijani. The Xe-133 and Xe-135 resulting from decays of the I-133 and I-135 by July 15 are included in those isotopes' inventories.

APPENDIX E REFERENCES

E1. Daniel & Associates, Inc., 136 Parker Road, Crescent City, Florida 32112-4736.

E2. "Plaintiffs' FRCP Rule 26(a)(2) Disclosure of Expert Testimony and Report of Arjun Makhijani," U. S. District Court, Central District of California, Western Division, Case No. 97-1554 DT (RCx), Lawrence O'Conner et al., Plaintiffs, v. Boeing North American, Inc., et al., Defendants (January 18, 2005).

F

Jerry D. Christian - Resume
3122 Homestead Lane
Idaho Falls, ID 83404-7195
Phone: (208)522-6793
FAX: (208)524-3561
E-mail: jerryc@srv.net

Scientific Fellow, Retired from Idaho National Engineering and Environmental Laboratory, September 2001; Consultant and President of Electrode Specialties Company
B. S. Chemistry, University of Oregon, 1959
Ph. D. Physical Chemistry, University of Washington, 1965
Postdoctoral: National Research Council Senior Research Associate, NASA Ames Research Center, Moffett Field, CA, 1972-1974
DOE "Q" (Secret) clearance reinstatement applied for March 2004, in process

INTERESTS: Provide consulting services on as-needed basis to develop and improve processes and technical concepts in areas of expertise – see Career Summary.

CAREER SUMMARY: I have thirty eight years experience in nuclear waste and fuel processing research and development. Included in achievements is development of the highly successful classified Fluorinel Dissolution Process for advanced naval fuels that was implemented in a new $250 million facility at the ICPP in the mid-80s. Career interests and accomplishments have been in the areas of nuclear fuel dissolution, aqueous fluoride chemistry, metal halide vaporization processes, high temperature ruthenium chemistry, solvent extraction separations chemistry, radioactive incinerator off-gas treatment, radioactive airborne waste management technologies, beneficial reuse of radioactive contaminated metals, oxidation of metals, thermodynamic modeling, activity coefficient calculations and models, technetium-molybdenum separations processes for medical isotope production, and characterization and simulation of radioactive sodium-bearing waste. Groundbreaking work on ruthenium chemistry changed prevailing concepts about its behavior and chemistry during evaporation and calcination of high-level waste solutions and has influenced researchers around the world.

I received a Special Award from the Chairman of Westinghouse for development of the Fluorinel Dissolution Process for advanced naval fuels, was one of ten annual international Westinghouse Signature Award winners in 1993 – for a chloride volatility processing concept for spent nuclear fuels – and received the 1994 ANS Special Award for Innovations in Long-Term Storage of Spent Nuclear Fuels. The Idaho Academy of Science awarded me their inaugural Distinguished Scientist/Engineer Award in 2000.

EXPERIENCE:

October 2001 to present — *Retired INEEL Scientific Fellow. Consultant; Manufacture electrode for measurement of free hydrofluoric acid.* Projects have included:

- Savannah River Laboratory: Design of experiments and data reduction to measure activity coefficients in process solutions to aid in modeling conditions to prevent precipitations.
- INEEL: Thermodynamic modeling of chemical composition of calcine for characterization purposes related to disposal of calcine in Yucca Mountain. Extensive use and expertise in HSC thermodynamic program and database.
- INEEL: Radiolysis modeling of HLLW.
- General: Referee for Nuclear Technology and Talanta journals; review

proposals for NEER program and for EMSP INEEL submittals.

- Manufacturing: developed and provided commercial instruments for measuring free HF in acid systems to BWXT fuel processing and fuel manufacturing plants and Savannah River Laboratory. BWXT has used the probe to correlate uranium extractability to free HF and improve system and as a safety control in fuel manufacturing chemical makeup.

Oct 1978 to
Sept 2001
and
Mar 1974 to
Jan 1977

Bechtel BWXT Idaho, LLC and predecessor DOE/ERDA/AEC prime contractors at the Idaho Chemical Processing Plant and INEEL, Idaho Falls, Idaho
Last Position: Scientific Fellow in Applied Technology Department.
Scientific Fellow is highest achievable technical ladder position at INEEL. Charter member, appointed in January 1987.

- Member of Technical Advisory Panel to INEEL Laboratory Director.
- Developed concept for separating lanthanides from spent nuclear fuel to enhance neutronics and economics of dry fuel recycle process.
- Directed development of efficient separations and concentration process for technetium-99m from molybdenum-99 that will enable production of the major medical imaging isotope by accelerator production of low specific activity molybdenum-99.
- Led in analysis of solvent extraction data for radionuclide separations from ICPP high activity wastes that resulted in new models that will have predictive capabilities necessary for engineering applications. New modeling concepts were developed that will result in improvements to other major extraction processes in the nuclear industry.
- Developed conceptual flowsheet for chloride volatility dry processing of a variety of DOE spent nuclear fuels to condition them for disposal. *This resulted in a prestigious George Westinghouse Signature Award of Excellence at the Corporate top ten level in 1994.*
- Developed a non-hazardous catalyst to replace mercuric nitrate for dissolution of spent aluminum nuclear fuels in nitric acid. *This resulted in a George Westinghouse Innovation Award at the Business Unit level in 1993, and is the subject of a patent.*
- Conceived the recycling of radioactive scrap metal from decommissioned reactors into dry spent fuel storage canisters. *This resulted in receiving the 1994 American Nuclear Society Special Award for Innovations in Long-Term Storage of Spent Nuclear Fuels.*
- Developed and presented an invited two week IAEA workshop on monitoring and management of radioactive effluents from nuclear power reactors at the Qinshan, China nuclear power reactor site.
- Developed and implemented a classified dissolution process for advanced Navy nuclear fuels in the DOE national number one priority new $250 million facility. *In less than one year, solved numerous technical problems that had been unsuccessfully addressed by others over a period of 16 years in areas of materials and corrosion, criticality safety control, precipitate control, and uranium dissolution effectiveness.* This process was essential to the continued receipt of Navy fuels and maintenance of the Navy nuclear fleet. *It resulted in a Westinghouse Special Award from the Chairman of Westinghouse.*

- Developed an electrochemical probe for measuring hydrofluoric acid in process solutions and directed research using it to simultaneously determine six zirconium fluoride complexation constants in acid systems. *This was key to the development and control of the Navy Zircaloy fuel dissolution process.* The probe is now available as a commercial instrument.
- Developed a sulfuric acid dissolution process and flowsheet for stainless steel in nuclear fuels in the presence of nitrates, which passivates stainless steel. *This was a vital flowsheet for the Navy fuel dissolution.*
- Developed with two colleagues, as DOE's Lead Lab for Airborne Wastes, the U. S. policy and strategy and managed DOE programs for radioactive airborne waste management, managing $34 million of R&D funds. *This was the most successful and cost-effective of DOE's waste management programs and the only one to establish and meet a completion schedule.*
- Developed design concept for off-gas treatment of proposed DOE slagging pyrolysis incinerator for radioactive wastes. This led to an invitation to write an International Atomic Energy Agency Technical Report on off-gas treatment.
- Developed dynamic dissolution model for uranium aluminide fuels and applied results to establish process controls to assure criticality safety in the process.
- Performed fundamental and applied research characterizing vaporization processes of ruthenium during calcination and wrote an invited definitive treatise on the behavior and control of ruthenium during waste solidification processes, which has guided others in the field internationally.

Jan 1977 to Oct 1978 — *Science Applications, Inc. (now SAIC)*, San Diego, California
Program Manager, Effluent Development

- Made extensive contributions to Generic Environmental Impact Statement for Radioactive Waste Isolation in Geologic Formation, largely in areas of **accident** analysis. Contributed to safety analysis report for Hanford radioactive melter.
- Developed off-gas treatment systems for nuclear fuels dissolution and for radioactive slagging pyrolysis incinerator.
- Wrote critical review on treatment methods for airborne effluents from high-level waste solidification processes.

Jan 1972 to March 1974 — *National Research Council Senior Postdoctoral Associate, NASA Ames Research Center*, Moffett Field, California

- Studied materials for Space Shuttle and showed that the nickel-based materials would reach an oxidation rate in reactive atomic oxygen in the upper atmosphere that was limited by saturation of metal ion defects, a newly discovered concept in oxidation theory.

Aug 1971 to Jan 1972 — *Westinghouse Hanford Co.*, Richland, Washington
Senior Research Scientist

- Provided early empirical and theoretical definition of the acid digestion process for treatment of radioactive wastes.
- Derived irreversible thermodynamic model for constituents in liquid sodium in the FFTF to develop an understanding and prediction of transport processes.

Aug 1965 to Aug 1971 *Battelle Northwest Laboratories*, Richland, Washington
Senior Research Scientist; Technical Leader; Manager of Fundamental Research Section

- Investigated thermodynamics of molten salts and of actinides at high temperatures.
- Directed AEC fundamental research program on actinide element chemistry.

PATENTS:

1. J. D. Christian and P. A. Anderson, "Mercury-Free Dissolution of Aluminum-Clad Fuel in Nitric Acid," U.S. Patent No. 5,364,603, November 15, 1994.
2. J. D. Christian, T. R. Thomas, and G. F. Kessinger, "Dry Halide Method for Separating the Components of Spent Nuclear Fuels," U.S. Patent No. 5,774,815, June 30, 1998.
3. R. G. Bennett, J. D. Christian, S. B. Grover, D. A. Petti, W. K. Terry, and W. Y. Yoon, "Method for Production of ^{99m}Tc Compositions from ^{99}Mo-Containing Materials," U.S. Patent No. 5,802,439, September 1, 1998.
4. R. G. Bennett, J. D. Christian, R. J. Kirkham, and T. J. Tranter, "Method for Generating a Crystalline $^{99}MoO_3$ Product and the Isolation of ^{99m}Tc Compositions Therefrom," U.S. Patent No. 5,802,438, September 1, 1998.
5. J. D. Christian and T. J. Tranter, "Method for Separating Sodium from Molybdenum in Aqueous Solutions on Acidic Alumina Column," Invention Disclosure, August 18, 1998.
6. J. W. Sterbentz and J. D. Christian, "Dry Processing Methods for Separation of Rare Earth Fission Products from Spent Nuclear Fuel," Invention Disclosure No. B-011, November 12, 1999.
7. J. D. Christian, P. A. Anderson, and K. N. Brewer, "Processing of Irradiated Stainless Steel to Reduce the Volume of Greater-than-Class C Waste," B-078 application submitted to DOE patent office October 3, 2000; DOE Case Number S-96,868.

PUBLICATIONS:

Peer-Reviewed Journals

1. J. D. Christian, A. A. Passchier and N. W. Gregory, "The Ultraviolet-Visible Absorption Spectrum of Bromine Between Room Temperature and 440 ," *J. Phys. Chem.*, **71**, 937-942 (1967). *Identified new Br_4 gas species.*
2. J. D. Christian and N. W. Gregory, "Vapor Phase Absorption Characteristics of Iron (III) Bromide and Iron (III) Chloride in the Ultraviolet-Visible Region," *J. Phys. Chem.*, **71**, 1579-1583 (1967). *Identified and characterized new dimeric iron (III) halide species.*
3. J. D. Christian and N. W. Gregory, "Equilibrium between Iron (III) Oxide and Hydrogen Bromide," *J. Phys. Chem.*, **71**, 1583-1587 (1967).
4. J. D. Christian, "The Strength of Chemical Bonds," *J. Chem. Educ.*, **50**, 176-177, 1973. *Presented new concept in depicting bond strength.*
5. J. D. Christian, "The Measurement of Gas Quantities by Liquid Displacement," *Anal. Chem.*, **45**, 698-702 (1973). *Increased state-of-the art accuracy by developing rigorous theoretical corrections and refinements to gas volume measurements and provided basis for apparatus design.*
6. J. D. Christian and W. P. Gilbreath, "Sealing of Large Leaks in High Vacuum Systems Subject to Thermal and Mechanical Stresses," *J. Vac. Sci. & Technol.*, **11**, 848 (1974). *New technique with important applications in vacuum technology. This paper received award from Amer. Vac. Soc. as Best Shop Note 1974.*
7. J. D. Christian and W. P. Gilbreath, "Apparatus for Production, Measurement and Reaction Studies of Dissociated Gases at Elevated Temperatures," *IEEE Trans. Plasma Science*, **PS-3**(2), 88-89 (1975). *New technology for atomic oxygen chemistry studies.*
8. J. D. Christian and W. P. Gilbreath, "Defect Structure of NiO and Rates and Mechanisms of Formation from Atomic Oxygen and Nickel," *Oxid. of Metals*, **9**, 1-25 (1975). *Discovered new concept of saturated surface Ni vacancies, limiting oxidation rate in upper atmosphere reentry of space shuttle.*
9. J. D. Christian, D. B. Illum, and J. A. Murphy, "Metal Electrodes for Continuous Amperometric Measurement of Free Hydrofluoric Acid in Acidic Solutions Containing Complexing Ions," *Talanta*,

37, 651-654 (1990). *Significant advance as tool for characterizing aqueous fluoride chemistry - technology has been transferred to BXW Technologies for processing Russian Sapphire fuel in DOE program.*

10. D. A. Knecht, M. D. Staiger, J. D. Christian, C. L. Bendixsen, G. W. Hogg, and J. R. Berreth, "Historical Fuel Reprocessing and HLW Management at Idaho," *Radwaste Magazine*, 35-47 (May 1997).
11. P. A. Anderson and J. D. Christian, "Alternative Reagent to Mercuric Nitrate Catalyst for Dissolution of Aluminum-Clad Nuclear Fuels in Nitric Acid," *J. Matls. Res.*, **13**, 68-76 (January 1998). *Special issue on environmentally benign materials and processes.*
12. K. N. Brewer, R. S. Herbst, T. A. Todd, and J. D. Christian, "Zirconium Extraction into Octyl(Phenyl)-N,N-Diisobutylcarbamoylmethyl Phosphine Oxide and Tributyl Phosphate," *Solv. Extract. & Ion Exchange*, **16**(4), 1047-1066 (1998).
13. D. A. Knecht, M. D. Staiger, J. D. Christian, and C. L. Bendixsen, "Historical Reprocessing of Spent Fuel and HLW Management at the Idaho Chemical Processing Plant," *Ceramic Transactions*, **87**, 177-186 (1998).
14. R. G. Bennett, J. D. Christian, D. A. Petti, W. K. Terry, and S. B. Grover, "A System of Technetium-99m Production Based on Distributed Electron Accelerators and Thermal Separation," *Nuclear Technology*, **126**, 102-121 (April 1999). This paper received referee comment that it was the "*one of the best written papers I have reviewed... Technical approach is outstanding.*
15. J. D. Christian, D. A. Petti, R. J. Kirkham, and R. G. Bennett, "Advances in Sublimation Separation of Technetium from Low-Specific-Activity Molybdenum-99," *Ind. & Eng. Chem. Res.*, **39**, 3157-3168 (September 2000).
16. J. D. Christian and I. R. Thomas, "Error Analysis of Uranium, Transuranium, and Neodymium Recoveries from Irradiated S3G-3 Fuels in the Fluorinel Dissolution Process at the Idaho Chemical Processing Plant," INEEL/EXT-2000-00113, Confidential-Restricted Data Report to Bettis Atomic Power Laboratory, November 2000. *This extensive report that includes process chemistry, calibration, sampling and measurement methodologies, and analytical methods, was thoroughly reviewed and refereed by Bettis Atomic Power Laboratory and Knolls Atomic Power Laboratory.*
17. J. D. Christian and W. J. Crooks III, "Densities of Sodium Tetrafluoroborate Aqueous Solutions at 20°C," *Indian J. Chem.*, in press.
18. J. A. Murphy and J. D. Christian, "Zirconium Fluoride Stability Constants Determined by Direct Amperometric Measurement of Equilibrium Hydrofluoric Acid," manuscript in preparation for submittal to *J. Inorg. Nucl. Chem.*
19. J. D. Christian, "An Improved Method of Precise Temperature Control of Insulated Electric Furnaces," manuscript prepared for submittal to *Rev. Sci. Instr.*
20. W. J. Crooks III, J. D. Christian, and W. D. Rhodes, "Thermodynamics of Saturated Aqueous Solutions of KBF_4 in Mixtures with $NaNO_3$, $NaBF_4$, and KNO_3" Manuscript in preparation for submittal to *Radiochimica Acta*.
21. W. J. Crooks III, J. D. Christian, and W. D. Rhodes, "Modeling of Solution Stabilities in Acidic Fluoride Nuclear Fuel Dissolution Solutions" Manuscript in preparation for submittal to *Nuclear Technology*.

Peer-Reviewed Conference Full Papers Published

1. J. D. Christian, "Behavior and Control of Ruthenium and Cerium," Proceedings of ANS-AIChE Topical Meeting on Controlling Airborne Effluents from Fuel Cycle Plants, Sun Valley, Idaho, August 5-6, 1976, 34 pp (*invited paper*). *Key paper in ruthenium literature, highly referenced. Described original studies showing that past concepts of the mechanism of ruthenium vaporization behavior were incorrect, identified new vapor species involved in calcination processes and showed how volatilization can be controlled. Also a comprehensive review of ruthenium and cerium*

literature and chemistry. Provided basis for subsequent IAEA Report Series No. 20, "Control of Semivolatile Radionuclides at Nuclear Facilities (1982).

2. D. W. Rhodes, W. A. Freeby, and J. D. Christian, "Ruthenium Containment during Fluidized-Bed Calcination of High-Level Waste," *AIChE Symposium Series*, 75, No. 191, 69-75 (1979).
3. J. D. Christian, "Zircaloy Corrosion in a Repository Environment," in Proceedings of Symposium on Scientific Basis for Nuclear Waste Management XXII, Boston, MA, Materials Research Society (1998), Pittsburgh, Published 1999.
4. J. D. Christian, J. W. Sterbentz, D. G. Abbott (INEEL), K. R. Czerwinski (Massachusetts Institute of Technology), and R. J. Cacciapouti (Duke Engineering & Services), "Concepts for Dry Processing of Spent Nuclear Fuel for Recycling to Light-Water Reactors," in Proceedings of Symposium on Scientific Basis for Nuclear Waste Management XXIII, Boston, MA, Materials Research Society (December 1999).
5. W. D. Rhodes, W. J. Crooks III, and J. D. Christian, "Use of Modeling for Prevention of Solids Formation During Canyon Processing of Legacy Nuclear Materials at the Savannah River Site," Proceedings WM'02 Conference, February 24-28, 2002, Tucson, AZ.
6. J. D. Christian and C. M. Frazee, "Thermodynamic Modeling of the Chemical Composition of Calcine at the Idaho Nuclear Technology and Engineering Center," Proceedings WM '04 Conference, February 29-March 4, 2004, Tucson, Arizona.

Books and Book Chapters

1. J. D. Christian, "Ruthenium," Chapter in *Technical Alternatives Document*, ERDA-76-43, May 1976.
2. J. D. Christian, A. Chrubasik, and P. Patek, "Treatment of Off-Gas from Radioactive Waste Incinerators," *IAEA (International Atomic Energy Agency) Technical Report, Series No. 302* (book), 1989. *Invited contribution as consultant to IAEA. Comprehensive assessment and recommendations for countries entering the field.*
3. J. D. Christian and T. R. Thomas, "Treatment of Off Gases from Process Operations," Chapter in *Separations Techniques in Nuclear Waste Management,* T. Carleson, C. Wai, and N. A. Chipman, editors, CRC Press, Inc., Cleveland, November 1995 (invited contribution).
4. J. D. Christian, "Fluidized-Bed Calcination," Chapter in *Hazardous and Radioactive Waste Treatment Technologies Handbook,* C. H. Oh, editor, CRC Press, Inc., Cleveland, June 2001 (invited contribution).

Key Documents and Reports

1. J. D. Christian, "A Spectrophotometric Study of Halide Vapor Phase Equilibria at Elevated Temperatures", Ph.D. Thesis, University of Washington (1965), University Microfilms Publication No. 66-5850, Ann Arbor, Michigan (165 + vii pages). *Developed new technique for studying thermodynamics of vaporization processes.*
2. J. D. Christian, "Summary Report of Acid Digestion Studies," report to Westinghouse Hanford Co., 1972. *Technical basis for process flowsheet and design of pilot plant demonstration facility for processing transuranic-contaminated combustible wastes.*
3. J. D. Christian and D. W. Rhodes, "Ruthenium Containment during Fluid-Bed Calcination of High Level Waste from Commercial Nuclear Fuel Reprocessing Plants," ICP-1091 (August 1976).
4. J. D. Christian and D. T. Pence, "Critical Methods of Treating Airborne Effluents from High-Level Waste Solidification Processes," PNL-2486 (June 1977). *Along with Conference Paper No. 1, above, this provided the basis for the International Atomic Energy Agency Technical Report Series No. 20, "Control of Semivolatile Radionuclides at Nuclear Facilities" (1982)*

5. J. D. Christian et al., "R&D for Off-Gas Treatment System for a Slagging Pyrolysis Radioactive Waste Incinerator - Final Report for Phase II," Science Applications, Inc. Report SAI78-904-LJ (October 27, 1978).
6. J. D. Christian et al., "Recommended Off-Gas Treatment System for the INEL Radioactive Waste Slagging Pyrolysis Incinerator," EG&G Idaho, Inc. Report PR-W-79-023 (August 1979). *Report of International Panel, J. D. Christian, Chairman. Important in-depth assessment of integrated system technology.*
7. R. A. Brown, J. D. Christian, and T. R. Thomas, "Airborne Radionuclide Waste Management," ENICO-1132 (June 1983). *Comprehensive compilation and analysis, along with reference 8, of source terms and dose impacts of airborne radionuclides from fuel reprocessing with recommended controls. Used to establish the U.S. strategy for airborne waste managements.*
8. R. A Brown, J. D. Christian, and T. R. Thomas, "Airborne Radionuclide Waste Management Reference Document," ENICO-1133 (July 1993).
9. J. D. Christian, T. R. Thomas, and G. F. Kessinger, "Pyrochemical Processing of Spent Nuclear Fuels," in *Potential Dispositioning Flowsheets for ICPP SNF and Wastes,"* A. L. Olson, editor, INEL-95/054, November 1995.
10. J. D. Christian and K. N. Brewer, "Processing of Metal and Calcined Sapphire Material," report prepared for Babcock and Wilcox Naval Nuclear Fuel Division, March 13, 1997. *Provided chemical basis for acidic fluoride processing of the Russian U-Be fuel, based on INEEL technologies developed for processing naval fuels.*
11. J. D. Christian, "Dissolution of Sand, Slag, and Crucible Residues," report prepared for Westinghouse Savannah River Company, January 5, 1998. *Provided chemical basis for acidic fluoride processing of the Rocky Flats residues, based on INEEL technologies developed for processing naval fuels.*
12. J. D. Christian, "Processing of Neptunium Targets for Pu-238 Recovery: Dissolution Chemistry," in Plutonium-238 Draft Environmental Impact Statement, May 1999. *This flowsheet for processing targets in the FDP facility quickly helped raise INEEL to a favored option from not being considered for processing the targets. The flowsheet provides a new chemical basis for dissolving the irradiated targets and is a major improvement over other site-proposed flowsheets; it incorporated an INEEL-patented fluoboric acid catalyst for dissolving aluminum and neptunium/plutonium oxides.*
13. J. D. Christian, J. W. Sterbentz, D. A. Abbott, D. A. Knecht, I. R. Thomas, N. E. Stanley, C. S. Staley, M. L. Croson (INEEL), R. J. Cacciapouti, G. M. Solan, M. C. Beganski, A. P. Fyfe (Duke Engineering & Services), K. R. Czerwinski, M. P. Reynard (Massachusetts Institute of Technology), and K. P. Carney (Argonne National Laboratory-West), "Dry Recycle of Spent Nuclear Fuel," J. D. Christian, Editor, INEEL LDRD Project 99-292 Final Report, Spring 2000, 185 + xi pages.
14. J. D. Christian and I. R. Thomas, "Error Analysis of Uranium, Transuranium, and Neodymium Recoveries from Irradiated S3G-3 Fuels in the Fluorinel Dissolution Process at the Idaho Chemical Processing Plant," INEEL/EXT-2000-00113, Confidential-Restricted Data Report to Bettis Atomic Power Laboratory, November 2000. (Requires DOE 'L' Clearance and need to know.) Provides detailed information on the dissolution process. Available by contacting the authors: J. D. Christian (208)522-6793; I. R. Thomas (208)526-3774.
15. J. D. Christian, "Composition and Simulation of Tank WM-180 Sodium-Bearing Waste at the Idaho Nuclear Technology and Engineering Center," INEEL/EXT-2001-00600, May 2001.
16. J. D. Christian, "'Worst-Case' Simulant for INTEC Sodium-Bearing Waste Vitrification Tests," INEEL/EXT-01-01219, September 2001.
17. W. J. Crooks III, W. D. Rhodes, and J. D. Christian, "Use of Modeling for the Prevention of Solids Formation during Canyon Processing of Legacy Nuclear Materials," WSRC-TR-00462, February 2003.
18. J. D. Christian, "Thermodynamic Modeling of Calcine Chemical Composition in CSSF II Bin 7 at INTEC," INEEL Engineering Design File, EDF-4059, December 15, 2003.

19. J. D. Christian, “Thermodynamic Modeling of the Chemical Composition for the NWCF H-3 Calcine Sample,” INEEL Engineering Design File, EDF-4556, March 2004.
20. J. D. Christian, “Thermodynamic Modeling of Calcine Chemical Composition in CSSF II Bin 3 at INTEC,” INEEL Engineering Design File, EDF-4XXX, April 2004.

KEY TECHNICAL PRESENTATIONS:

1. J. D. Christian, “Vaporization of Uranium Trichloride: Evidence for the Formation of $UCl_2(c)$,” paper presented at 23rd NW Regional Meeting of the American Chemical Society, June 12-14, 1968, Portland, Oregon. *Identified and determined thermodynamics of new species of* UCl_2.
2. J. D. Christian, “The Enthalpy of UAl_3 from 295 to 1200K,” paper presented at 24th Calorimetry Conference, Portsmouth, New Hampshire, October 1969.
3. J. D. Christian and W. P. Gilbreath, “The Rates and Mechanism of Nickel Oxidation by Atomic Oxygen,” paper presented at International Symposium on Plasma Chemistry, Kiel, Germany, September 1973.
4. D. T. Pence, J. D. Christian, and W. J. Paplawsky, “Noble Gas Separation with the Use of Inorganic Adsorbents,” Proceedings of 15th DOE Nuclear Air Cleaning Conference, San Diego, California, 1979. *New concept for gas separations. Included ruthenium removal in catalytic bed for* NH_3 *reduction of* NO_x.
7. J. D. Christian et al., “Off-Gas Treatment for INEL Radioactive Slagging Pyrolysis Incinerator,” Waste Management ‘80, Proceedings of Symposium on Waste Management at Tucson, Arizona, March 1980 (*Invited paper*).
8. J. D. Christian, “Analysis of Airborne Waste Management Problems with Recommendations for Control Standards,” paper presented at Workshop on Dissolver Off-Gas Treatment, Karlsruhe, Federal Republic of Germany, April 1982.
9. R. A. Brown, J. D. Christian, and T. R. Thomas, “Recovery and Storage Policy Decisions for Airborne Radionuclides,” Proceedings of International Conference on Radioactive Waste Management held by the International Atomic Energy Agency, Seattle, Washington, May 16-20, 1983 (*invited paper*).
10. R. A. Brown, J. D. Christian, and T. R. Thomas, “The Status of Development of U.S. Technologies for the Management of Airborne Radionuclides,” paper presented at U.S. DOE and French Commissariat A L’Energie Atomique Meeting on Radionuclide Waste Management, Knoxville, Tennessee, November 9-11, 1983.
11. J. F. Krupa, J. D. Christian, and S. D. Reeder, “Control of Zircaloy Fuel Dissolution Chemistry Using an n-Silicon Electrode,” Transactions of American Nuclear Society 1985 Summer Meeting, Boston, Massachusetts, June 9-14, 1985. *Innovative method for determining HF complexing needs in system involving multiple equilibria to control the HF concentration at elevated temperatures. Crucial for operation of new Fluorinel dissolution process.*
12. J. D. Christian and P. A. Anderson, “Dissolution of Stainless Steel in Sulfuric Acid in Presence of Nitric Acid Residue,” paper presented at 41st NW Regional Meeting of the American Chemical Society, Portland, Oregon, June 9-16, 1986.
13. J. D. Christian, “Process Control During Dissolution of Zircaloy-Based Fuels,” paper presented at 196th ACS National Meeting, Los Angeles, California, September 25-39 (1989). (*Invited paper*).
14. J. D. Christian and J. A. Murphy, “Process Analysis for Controlling and Improving Nuclear Fuel Dissolution,’ paper presented at Symposium on New Developments in Process Analysis, 44th NW Regional Meeting of the American Chemical Society, Reno, Nevada, June 14-16, 1989 (*invited paper*).
15. D. R. Evans, J. D. Christian, and R. F. Farman, “Simulation of Uranium Aluminide Dissolution in a Continuous Aluminum Dissolver System,” Proceedings of Summer Computer Simulation conference ‘90, Calgary, Alberta, Canada, July 1990.

16. J. D. Christian, "Behavior and Control of Ruthenium During Operation of the New Waste Calcining Facility at the Idaho Chemical Processing Plant," Proceedings of 21st DOE/NRC Nuclear Air Cleaning Conference, San Diego, California, August 13-17, 1990.
17. J. D. Christian and R. A. Nelson, "Monitoring and Management of Radioactive Effluents from Nuclear Power Reactors," two week IAEA workshop presented at Qinshan, China nuclear power reactor site for the China National Nuclear Corp., December 3-14, 1990.
18. P. A. Anderson and J. D. Christian, "Mercury-Free Dissolution of Aluminum Fuels in Nitric Acid," Trans. Amer. Nuclear Soc., 69, 1993 (Presented at San Francisco meeting of ANS, November 14-18, 1993).
19. J. D. Christian, T. R. Thomas, and G. F. Kessinger, "A Dry Chloride Volatility Concept for Processing Spent Nuclear Fuels," Proceedings of Spectrum '94, International Topical Meeting on Nuclear and Hazardous Waste Management (American Nuclear Society Fuel Cycle Division), Atlanta, Georgia, August 14-18, 1994.
20. J. D. Christian, "Fabrication of Multi-Purpose Canisters from Radioactive Scrap Metal," Proceedings of DOE Spent Nuclear Fuel, Challenges & Initiatives, Salt Lake City, December 13-16, 1994.
21. K. N. Brewer, R. S. Herbst, J. D. Christian, and T. A. Todd, "Zirconium Chemistry in the TRUEX Process Solvent," Proceedings of Actinide Separations Conference, Monterey, California, June 12-16, 1995.
22. J. D. Christian, D. A. Petti, and R. J. Kirkham, "A Distributed Production System for ^{99m}Tc. II. Separation Processing," Trans. Amer. Nuclear Soc.,77, 557 (1997).
23. J. D. Christian, J. W. Sterbentz, D. G. Abbott (INEEL), R. J. Cacciaputi (Duke Engineering Services), and K. R. Czerwinski (MIT), "Research on Dry Processing for Recycle of Spent Nuclear Fuel," Proceedings of GLOBAL '99 International Conference on Future Nuclear Systems, August 29-September 3, 1999, Jackson Hole, Wyoming.
24. R. G. Bennett, W. K. Terry, J. D. Christian, R. J. Kirkham, and D. A. Petti, "A Practical Example of ^{99m}Tc Distribution Based on Electron Accelerator Production," Proceedings of GLOBAL '99 International Conference on Future Nuclear Systems, August 29-September 3, 1999, Jackson Hole, Wyoming.
25. J. D. Christian, "Aqueous Fluoride Chemistry in Nuclear Fuel Dissolution," Proceedings Idaho Academy of Science Annual Meeting, March 2000 (abstract published). Also, invited lecture presented to Idaho State Graduate Engineering Department, April 12, 2000.
26. J. D. Christian, P. A. Anderson, and K. N. Brewer, "Processing of Irradiated Stainless Steel for Reduction of GTCC Waste," Proceedings of Spectrum 2000, International Conference on Nuclear and Hazardous Waste Management, September 24-28, 2000, Chattanooga, TN.
27. J. D. Christian, "Fuel Reprocessing at the Idaho National Engineering and Environmental Laboratory – Fuel Dissolution and Wastes," invited paper presented at Symposium on Radionuclide Site Remediation, 58th NW Regional Meeting of the American Chemical Society, June 12-14, 2003, Bozeman, Montana (abstract published).

KEY INTERNAL LETTER REPORTS:

Much of work and original research performed since May 1983 on Navy fuels is classified and, thus, not publishable. Therefore, results are documented in internal confidential-restricted data letters. The original research results had major impact on successful implementation of a new process. Other technical and innovative work is of such a nature that it is documented by letter reports.

J. D. Christian, letter (Westinghouse Idaho Nuclear Co. and predecessor DOE contractor, Exxon Idaho Nuclear Co.):

1. JDC-4-75 to D. W. Rhodes, "Additives to Increase Calcine Attrition Resistance," April 14, 1975. *Key analysis that led to understanding and control of powder fines during production of calcine in Waste Calcining Facility.*
2. JDC-7-83 to D. L. Condotta, "Component Ratios in Fluorinel Fuels," April 14, 1983 (confidential-restricted data). *Resolved major error in understanding of fuel properties affecting processing that was leading unnecessarily to major plant modifications.*
3. JDC-10-84 to B. R. Dickey, "Fluorinel Fuel Dissolution and Uranium Heelout Flowsheets for FDP Startup," June 29, 1984 (confidential-restricted data). *Key document for startup operations of new plant.*
4. JDC-16-84 to R. A. Brown et al., "Suggested Criteria for Successful Fluorinel Uranium Heelout," November 30, 1984. *Provided the chemical basis for safety envelop that eliminated excess duplicate uranium heelout processing, saving $3.6 million annually.*
5. JDC-4-85 to B. R. Dickey, "Fluorinel Stainless Steel Heelout Flowsheet for FDP Startup," January 30, 1985.
6. JDC-19-85 to R. E. Mizia, "Design of Corrosion Tests for Fluorinel Uranium-Bearing Solids Dissolution Conditions," October 2, 1985.
7. JDC-20-85 to R. J. Bliss, "Results of FDP Dissolver K-Effective Modeling Studies - Deletion of Fluoboric Acid Addition Requirements," November 13, 1985 (confidential-restricted data). *Experimental and modeling basis for eliminating requirement for $2 million reagent addition system.*
8. JDC-21-85 to R. J. Bliss, "Flowsheet Changes for FDP Phase II Startup," December 23, 1985 (confidential-restricted data). *Key document for the second campaign operations - implemented process and safety control improvements as result of operational experience.*
9. JDC-1-86 to R. J. Bliss, "^{235}U Depletion Calculation for FDP Fuels: Report of Task Force," January 27, 1986. *Essential theoretical development that enabled required mass balance calculations for criticality safety in FDP operations for processing advanced naval fuels.*
10. JDC-9-86 to R. J. Bliss, "Stainless Steel Heelout Flowsheets for FDP Phase II," May 1986.
11. J. D. Christian to R. J. Bliss et al., "Summary of FDP Phase I Process Flowsheet Performance," September 11, 1986.
12. JDC-27-86 to W. B. Palmer, "Design of ^{137}Cs Spike Dissolution Experiments in FDP," September 24, 1986. *The first "hot" operating test in the new plant.*
13. JDC-1-87 to G. W. Hogg, "Preliminary FDP Upgrade Fuel Dissolution Flowsheet," January 14, 1987 (confidential-restricted data).
14. JDC-7-87 to B. K. Hampton, "New Uranium Heelout Flowsheet for Use in FDP," January 18, 1987.
15. JDC-17-87 to J. L. Lee, "Elimination of Stainless Steel Heelout in Irradiated Fluorinel Dissolution Process," April 23, 1987.
16. JDC-25-87 to J. A. Rindfleisch, "NO_X Control in FDP for Protection of Trickle-Bed Reactor," July 21, 1987.
17. JDC-38-87 to J. A. Rindfleisch, "Elimination of Zirconyl Nitrate from FDP Uranium Heelout Flowsheet," January 8, 1988.
18. RID-8-88/JDC-8-88 (with R. I. Donovan), "Irradiated Fuel Dissolution Test - Run 46/Phase V," May 26, 1988 (confidential-restricted data).
19. JDC-16-88 to M. D. Staiger, "Preliminary Fuel Dissolution Flowsheet for FDP Geometrically Favorable Dissolver," June 30, 1988 (confidential-restricted data).
20. JDC-28-88 to G. T. Paulson, "Analyzed Compositions of FDP Bulk Uranium-Bearing Solids from PTV," July 10, 1988 (confidential-restricted data). *Thorough assessment; results used extensively in criticality safety controls and design of new high level waste tanks.*
21. JDC-24-88 to B. R. Wheeler, "Technical Basis for Dissolution of NWCF Bed on October 31, 1988," October 31, 1988. *Chemistry controls to prevent Ru-106 releases.*

22. JDC-16-89/RFF-6-89 (with R. F. Farman) to J. E. Johnson, "Process Model for Uranium Aluminide Fuel Dissolution During Continuous Al Fuel Processing in G-Cell Dissolvers," August 12, 1989. *Along with 23 below provided criticality safety envelope basis for planned campaign.*
23. JDC-21-89/DRE-7-89 (with D. R. Evans) to J. E. Johnson, "Process Control Limits for Continuous Al Fuel Dissolution System," August 21, 1989.
24. JDC-30-89 to C. D. Griffin, "FDP Empirical Equations for FHU Contents," November 13, 1989 (confidential-restricted data). *Technical basis for implementing new criticality safety controls that eliminated need for costly uranium heelouts - derived from operational data.*

PROFESSIONAL ACTIVITIES:

- University of Washington Joint Center for Graduate Study, Richland
  Lecturer in Chemistry (1965 to 1971).
- University of Idaho, Idaho Falls Center. Instructor (1987 to present).
- Appointed Affiliate Professor of Chemistry 1998.
- Treasurer of Idaho Section of American Chemical Society, 1976.
- General Chairman 40th NW Regional Meeting of American Chemical Society, 1985.
- Chairman-Elect 1986 and Chairman 1987 of Idaho Section of American Chemical Society.
- Thesis adviser for University of Idaho Master of Science degree in chemistry for three students, 1991 and 1992.
- Reviewer/referee for *Nuclear Technology*, a Journal of the American Nuclear Society, 1988 to present.
- Referee for *Talanta*, International Journal of Pure and Applied Analytical Chemistry, 1989 to present.

AWARDS OF EXTERNAL RECOGNITION:

- NASA Technology Utilization Award for Creative Development of method for accurate measurement of gas volumes by liquid displacement, 1973.
- American Vacuum Society Award for Best Shop Note in J. Vac. Science and Technol. in 1974.
- NASA Technology Utilization Award for Creative Development of Technology for development of method for semipermanent sealing of leaks in high vacuum systems subject to thermal and mechanical stresses, 1975.
- NASA Technology Utilization Award for Creative Development of Technology for development of apparatus for study of plasmas at elevated temperatures, 1976.
- Westinghouse Excellence Award from Advanced Power Systems Division, 1984.
- Westinghouse Engineering Achievement Award, 1985, for development of dissolution chemistry for special water reactor ternary oxide fuel.
- Westinghouse Special Award, and $2,500, 1985, from Chairman of Westinghouse, for development and implementation of Fluorinel fuel dissolution flowsheets for DOE's highest priority project.
- Westinghouse Innovation Business Award, 1993, for invention of fluoboric acid-catalyzed dissolution of aluminum alloy fuels in nitric acid.

- Westinghouse Signature Award of Excellence, 1993 winner (one of ten Corporate-wide), for development of dry chloride volatility processing concept for spent nuclear fuels.
- National 1994 American Nuclear Society Special Award, for Innovations in Long-Term Storage of Spent Nuclear Fuels, with $1000. One Special Award is given annually.
- Idaho Academy of Science inaugural Distinguished Scientist/Engineer Award, March 2000, "For an outstanding career of pioneering scientific research and leadership bringing recognition to Idaho science with national and international impact."

IMPACTS OF TECHNICAL ACHIEVEMENTS:

- Developments and expertise in aqueous fluoride chemistry (speciation of multiple fluoride species and invention of metal electrodes for amperometrically measuring free HF) have resulted in solutions to processing problems for naval nuclear fuels and in numerous requests for help in processing plants and in solvent extraction modeling.

INEEL

Advanced Naval Fuel Dissolution

- Problematic process taken to *DOE showcase* - most visible and important project in the complex
- Process implemented in new $250 M plant.
- Balanced fluoride chemistry - dissolution effectiveness; corrosion control; precipitate control

Continuous In-Line Complexing

- Upgrade of naval fuel process concept from batch to continuous: *improved throughput*
  - Utilizes titanium electrode and speciation calculations to measure and control F^- complexation
  - Invented HF electrode to monitor and control process

HBF_4 Catalyst for Al Fuel Dissolution

- Replaced mercuric nitrate, used since 1950's to dissolve fuels in nitric acid
- Saved $20 million – eliminated electrolytic dissolver project

TRUEX Separation of Radionuclides from Dissolved Calcine

- Explained observed Zr^{+4} 30% extraction in the face of 99.9999% Zr being complexed with fluoride.
- Guided development chemistry for separating Zr from transuranics.

Zirconium Fluoride Chemistry

- U of I Master's student under my direction as thesis adviser determined six sequential zirconium fluoride complexation constants using the metal electrode developed for measuring free HF. This vastly improved the values over those determined with a fluoride-selective electrode.

***Argonne National Laboratory* TRUEX Process**

- Incorporated zirconium fluoride complexation constants determined by the metal electrode into their Generalized TRUEX Model to improve ability to predict zirconium behavior during extraction from acidic fluoride wastes

Savannah River Site

Dissolution of Rocky Flats Sand-Slag-Crucible Plutonium Oxide Residues

- Modeled fluoride dissolution conditions to assure dissolution, prevent precipitation, and control corrosion
- Results implemented in plant processing

Dissolution of Aluminum Fuels

- Replaced mercuric nitrate with HBF4 catalyst developed in my laboratory with P. A. Anderson to dissolve aluminum-plutonium oxide fuels, starting January 2001.
- SRS is collaborating with Master's thesis student to study mechanism of the catalyst, using the metal electrodes developed in my laboratory for directly measuring free HF.

Solubility Products and Activity Coefficients of Potentially Precipitating Plant Species

- Presently developing activity coefficient models and thermodynamic speciation program and designing solubility product experiments to enable prediction and control of precipitating species in plant process multielectrolyte solutions.

BWX Technologies

Naval fuel dissolution:

- Using metal electrode system that I provided to monitor free HF in naval fuel dissolution process in their plant on a daily basis to improve conditions for uranium separation and recovery.
- Planning to implement continuous in-line complexing of fluoride developed at INTEC using metal electrodes for process control – major upgrade to process

Russian Sapphire fuel dissolution:

- Was asked to solve dissolution difficulties and developed a dissolution flowsheet for oxidized Russian U-Be Sapphire fuel. The flowsheet was used in successful processing operations.

Aluminum fuel dissolution:

- BWX Technologies is proposing dissolving aluminum fuels for uranium recovery using the patented HBF_4 catalyst developed by me (in collaboration with Mr. P. A. Anderson)

Advised *Russian Klophin Radium Institute* on Chemical Conditions for Universal Solvent Extraction of Transuranics from Dissolved Calcine

- Resulted in recommended changes to eliminate precipitates.

- Expertise in ruthenium chemistry has resulted in requests to solve operational problems at Hanford and to guide research studies in Japan and France.

Hanford tank ruthenium-106 releases

- Resolved problem of Ru-106 releases from high-level liquid waste tanks exceeding regulatory limits – explained observations and suggested practical fixes.

Hitachi Works, Japan

- Advised scientists studying ruthenium volatility from evaporating high-level liquid waste on scientific approaches for characterizing the ruthenium to address corrosion control in liquid waste evaporators in Japan's first commercial reprocessing plant.

REFERENCES:

1. Mr. Russell A. Brown (retired)
Former supervisor at Westinghouse Idaho Nuclear Co.
162 East 2nd Street
Idaho Falls, ID 83404
(208)524-4409
e-mail: rusb@qwest.net

2. Dr. William J. Crooks III
Los Alamos National Laboratory
MS E505
Los Alamos, NM 87545
(505)-665-8781
e-mail: crooks@lanl.gov

3. Dr. G. William Hogg (retired)
Former supervisor at Westinghouse Idaho Nuclear Co.
2940 Chaparral Drive
Idaho Falls, ID 83404
(208)523-4462
e-mail: tunimist@aol.com

4. Professor Jay Kunze
Dean, College of Engineering
Idaho State University
Campus Box 8060
Pocatello, ID 83209-8060
(208)236-2902
e-mail: kunzejay@isu.edu

5. Mr. Steve Lambson
Manager, Chemical Processing
Mail Stop 8193
BWXT Y-12
PO Box 2009
Oak Ridge, TN 37831
(865)576-4397
e-mail: lb8@Y12.doe.gov

6. Dr. Debbie Wachel
Knolls Atomic Power Laboratory
PO Box 7450, Bin No. 114
E6-211B
Schenectady, NY 12301
(518) 395-7075
e-mail: wachel@kapl.gov

7. Dr. William P. Gilbreath (retired from NASA)
928 Kings Blvd
Sun City, FL 33573
(813)634-4533
wgilbrea@tampabay.rr.com

8. Dr. Juergen Furrer
Institut für Teschnische Chemie
Bereich Thermische Abfallbehandlung
Forschungszentrum Karlsruhe GmbH
D-76021 Karlsruhe, Germany (retired)
e-mail: juergen.furrer@freenet.de

9. Mr. Russell A. Karl,* Dr. Jane Toben,
Dr. Mike Weisfield
Bettis Atomic Power Laboratory
P. O. Box 79
West Mifflin, PA 15122-0079
(412)476-5037; 476-6063; 476-5441
e-mail: tobenjm@bettis.gov
*Retired: 6122 Boxer Dr., Bethel Park, PA 15102; (412)833-4624

10. Dr. Ken Czerwinski
(Formerly Nuclear Engineering Dept., MIT)
Department of Chemistry
University of Nevada, Las Vegas
4305 Maryland Parkway
Las Vegas, NV 89154-4003
(702)895-0501
e-mail: czerwin2@unlv.nevada.edu

11. Dr. Valery Romanovsky
Deputy Director General
V. G. Khlopin Radium Institute
28, 2nd Murinskiy Ave.
St. Petersburg, 194021, Russia
e-mail: romanovski@atom.nw.ru

12. Mr. William C. Moffitt (retired)
Former President of Westinghouse Idaho Nuclear Company
731 Kohler Road
Pasco, WA 99301
(509) 544-0608
bjmoffitt1@msn.com

13. Dr. Billy R. Dickey (retired) Former Branch Manager and Vice President Technical Department Exxon Nuclear Idaho Company P. O. Box 901763 Sandy, UT 84090 (801)942-8502	14. Dr. Ralph G. Bennett Director, Advanced Nuclear Energy Idaho National Engineering & Environmental Laboratory P. O. Box 1625 Idaho Falls, ID 83415-3860 (208)526-7708 e-mail: rcb@inel.gov
15. Dr. Richard Boardman Consulting Engineer* Idaho National Engineering & Environmental Laboratory P. O. Box 1625 Idaho Falls, ID 83415-5218 (208)526-3732 e-mail: rdb1@inel.gov *Winner of Idaho Academy of Science Distinguished Scientist/Engineer Award, 2004	