Review and Evaluation of Report of David A Lochbaum

By

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My name is John Krsul, and I am an analytical chemist with more than forty years of experience in the preparation of analytical methods to support fabrication of advanced nuclear fuels, operation of an experimental breeder reactor, post-irradiation examination of reactor fuels, and the electrometallurgical treatment of spent reactor fuels. I was asked by The Boeing Company to review and comment on the report of David A. Lochbaum, entitled "An Assessment of Potential Pathways for Release of the Gaseous Radioactivity Following Fuel Damage During Run 14 at the Sodium Reactor Experiment," October 5, 2006 (identified as the "Lochbaum Report" for this review and evaluation).

My work has focused primarily on isotopic and elemental analyses of fission products, activation products, uranium and transuranium elements. I have published or co-authored more than thirty-five articles in the scientific literature, analytical reports, seminar papers, and a book chapter regarding my research and related interests. Before retiring, I worked for the Argonne National Laboratory ("ANL"), Nuclear Technology Division. My work at ANL primarily involved the Experimental Breeder Reactor II (the "EBR-II"). This reactor, now decommissioned, is located about 35 miles west of Idaho Falls, Idaho. EBR-II was a liquid metal fast breeder reactor. As part of my work on the EBR-II, I staffed and managed a high-quality analytical and radiochemical laboratory with broad capabilities in the support of ANL applied research projects. I provided supervision for the development of specialized wet-chemical, radiochemical, and instrumental methods of analysis, and provided chemical and radiochemical expertise to ANL project

personnel. I also proposed and coordinated a variety of experimental work, reviewed the experimented proposals of others, and reviewed designs for new and modified facilities for nuclear, environmental, and chemical compatibility and safety.

The EBR-II shared certain important similarities with the Sodium Reactor Experiment (the "SRE"), the reactor at issue in the Lochbaum Report. For example, for both the SRE and the EBR-II, the coolant was liquid sodium and the fuel was a metal. As is discussed below, the behavior and fate of fission products such as iodine-131 and cesium-137 at high temperatures in sodium-cooled, metal-fuel reactors like the SRE is very different from the behavior of these radionuclides in commercial nuclear power plant light water reactors in the United States, which use non-metal uranium dioxide fuel and are cooled with water. Another similarity between the EBR-II and SRE is that the cover gas used in the EBR-II was argon, an inert and very un-reactive gas similar to the helium used for the cover gas of the SRE.

As an analytical chemist at EBR-II, I was responsible for sampling and analyzing the EBR-II cover gas system, and the primary and secondary sodium systems. I was also responsible for the analysis of virgin and recycled reactor fuel samples, sampling and analysis of fuel recycling samples, sampling and analysis of spent fuel for post-irradiation examinations, including cladding and bond sodium, sampling and analyses for a variety of experiments, other site analytical chemistry requirements such as facility system monitoring, waste characterization and environmental samples. Particularly relevant for this analysis of the Lochbaum Report is my experience in sampling and analysis of cover gas and sodium coolant when fuel failed, when fuel was intentionally failed, and during run beyond cladding breach experiments.

To facilitate the review and evaluation of the Lochbaum Report, I reviewed certain documents regarding the SRE incident in 1959, and related subjects. Particularly important for this review are the following reports:

- *SRE Fuel Element Damage, Interim Report*, NAA-SR-4488, Atomics International, Nov. 15, 1959.

- *SRE Fuel Element Damage Final Report*, NAA-SR-4488(suppl), Atomics International, 1961.

- *Distribution of Fission Product Contamination in the SRE*, R.S. Hart, NAA-SR-6890, Atomics International, 1962.

- *Investigation of Releases from Santa Susana Sodium Reactor Experiment in July, 1959*, John A. Daniel, Sr., Daniel & Associates, Inc., May 27, 2005

- *Chemical Behavior of Iodine-131 during SRE Fuel Element Damage in July 1959*, Jerry D. Christian, May, 26, 2005.

Other references used are included at the end of this report.

Summary of Comments. The Lochbaum Report rejects previous conclusions that the 1959 SRE incident did not result in a radiological hazard to the reactor environs, and instead promotes a theory that up to 30 percent of the core inventory in the SRE was released to the atmosphere in the 1959 incident. This theory is based on the assumption that the gaseous radioactivity in 13 of the 43 fuel elements of the SRE was released to the atmosphere. The Lochbaum Report also endorses a "midpoint" estimate that 15 percent of the SRE failed fuel fission product inventory was released in the incident. Based on

my experience and knowledge, review of scientific literature, and the record relating to the 1959 SRE incident, I conclude that these qualitative release fractions stated in the Lochbaum Report are not scientifically supportable. As is discussed in more detail below, my opinion is based in part on the following facts:

- The Lochbaum Report ignores differences between the behavior of fission products in uranium metal fuel, such as that used in the SRE, and ceramic fuels (uranium oxide). Experiments with the EBR-II reactor shows that fission gases will not be released from uranium metal fuel until it has swelled to volumes corresponding to burnups of about 1%, while SRE uranium metal fuels were burned to 0.1% or less. Moreover, EBR-II experiments with failed uranium metal fuel, in temperatures higher than those experienced in the SRE incident, demonstrated that iodine did not vaporize as elemental iodine (as the Lochbaum Report assumes), but rather it acted like a metal iodide and was likely bound up with uranium in the fuel.
- The Lochbaum Report assumes, in both its "lower bound" and "upper bound" estimates, that any "radioiodine and other gases" released from the SRE fuel in the incident would have traveled unimpeded up through a six-foot deep (or deeper) sodium pool, reaching the cover gas where either all of the radioactivity (in the case of iodine-131) or between 10 and 100 percent of the radioactivity (in the case of cesium-137) would subsequently be released to the atmosphere. But this theory is not supported by any scientific literature, and ignores that according to basic laws of chemistry, elemental iodine and cesium would bond with the sodium coolant, preventing release of these radionuclides to the cover gas.

Summary of Review. The Lochbaum Report addresses issues regarding the SRE incident in July 1959, after which 13 of 43 SRE fuel rods showed some amount of damage.[1,2,3,4,5,6,7] The Lochbaum Report quotes some of the historical literature regarding

the incident to discuss issues such as: high temperatures in the primary sodium coolant, uncontrolled reactivity, coolant flow restrictions due to decomposition of tetralin, fuel failure, cover gas activities, gas storage tank activities, sodium sample activity, stack monitor activity, air monitoring activities, and radiation surveys. The Report also hypothesizes pathways to the release of fission products to the environment. The Report takes issue with (or at least cites as relevant) certain conclusions it attributes to Atomics International regarding the incident:

> *In spite of the cladding failures to at least 11 of the fuel elements, no radiological hazard was present to the reactor environs.*[1]

> *In spite of the cladding failure to 13 fuel elements and the release to the primary sodium of several thousand curies of fission product activity, no radiological hazard was presented to the reactor environs*.[2]

> *Considering a total accumulated burnup for the entire core 2250 Mwd, assuming that 1 Mw corresponds to a fission of 1 gm of material per day, and assuming the calculated average release fraction of about 4 x 10^{-4}, it is calculated that a total of 0.9 gm of fission products were released to the sodium. This corresponds to a concentration in sodium of only 0.04 ppm.*[1]

> *No iodine was ever detected in the reactor core gas samples.*[1]

> *Strontium-90 has deposited to the extent of about 0.8μCi/cm^2.* [1]

> *With the exception of the inert gases Xe133 and Kr85, all the fission fragments remained in the sodium, or were absorbed by the carbon or by the sodium-wetted metal surfaces.*[2]

As a result of the SRE fuel damage, reactor core gas activity was initially 10^{-3} μCi/cc, but after extensive purging plus normal decay, activity stabilized around 10^{-6} μCi/cc…Since these values greatly exceed tolerance levels, only a small amount of reactor cover gas could be allowed to pass into the working area.[5]

The cold trap located in the primary system was effective in removing fission product contamination.[3]

One would expect the release of iodine to be considerable higher than that of cesium rather than one-third of it as actually found. …A possible answer to the low iodine value would be the escape of the element from the primary sodium to the cover gas. However, iodine has never been found in the cover gas of the SRE either after this incident or in any of many other gas samples taken during the operation of the reactor.[3]

Although 5,000 to 10,000 curies of fission product activity were unexpectedly released to the primary sodium system no radiological emergency of any nature occurred.[3]

The main premise of the Lochbaum Report is that the company's claim that "no radiological emergency of any nature occurred"[3] "is not intuitively obvious given the fact that about one third (13 of 43 fuel elements) of the SRE reactor core was severely damaged and the reactor continued to operate for nearly two weeks after this damage likely occurred (July 13, 1959)". The Lochbaum Report rejects the conclusions of Atomics International ("AI") and alleges that all containment was breeched and radioiodine and cesium were released in large quantities to the atmosphere. In doing so,

Lochbaum describes two potential pathways to release radioactivity to the atmosphere. The pathways are through the radioactive gas storage tanks and the SRE building ventilation system. These proposed pathways are discussed below. Lochbaum also performs a "qualitative assessment," because he says that the available data does not permit a quantitative analysis.

To perform his qualitative assessment, Lochbaum says that he relies on information from fuel damage at the Enrico Fermi Atomic Power Station (or Fermi-1) in 1966 and from a literature search for experimental work/analyses. The Lochbaum Report ends with a conclusion that gives a lower and upper bounding condition for release for Cs-137 and I-131 as 0.3 to 30 percent and 3 to 30 percent, respectively. The bounding conditions are than attenuated by qualitative statements, noticeably without any data, that drive the lower bounding conditions and the upper bounding condition to "say 15 percent," as closer to the actual release fraction to the atmosphere.

Review and Evaluation of the Lochbaum Report.

This review evaluates the Lochbaum Report by comparing its qualitative assessments against recent reports, the original AI reports, my experience working at the EBR-II reactor with failed fuel and related experiments, and the scientific literature dealing specifically with the behavior of iodine and cesium in these types of systems.

This report is divided into five sections. Section 1 describes a number of reactor components, systems, and facilities pertinent to this review and evaluation. Section 2 reviews and evaluates information in reports that are specific to the failure of the fuel in July 1959. Section 3 reviews the Lochbaum qualitative assessment with respect to

radiological consequences. Section 4 reviews the Lochbaum section on release pathways. Section 5 reviews and evaluates the literature sited in the Lochbaum Report.

Section 1. Reactor components, systems, and facilities.

A number of reactor components, systems, and facilities are pertinent to this review. A brief description of each is given below. Significantly more detailed descriptions can be found in the literature.

The SRE was a sodium-cooled, graphite-moderated reactor using slightly enriched metal fuel. The reactor was encased in a primary sodium core tank. During operation of the reactor, liquid sodium circulated throughout (and above) the reactor core, serving as a coolant. The coolant has some containment properties. Heat from the reactor core, 6 feet in diameter and 6 feet high, was removed by a primary sodium system (radioactive). Heat wash transferred a secondary sodium system (non-radioactive). Sodium entered above the core through double-walled pipes. The pipes delivered the sodium to a plenum chamber at the base of the tank where the sodium was discharged. Sodium flowed up through channels provided in the moderator assemblies, removing heat from the suspended fuel elements. Sodium from the core flowed into a 6-foot deep pool, then to outlet pipes leading to the main and auxiliary primary loops and intermediate heat exchangers. A secondary sodium system delivered the heat to a generator to produce electricity or to an airblast heat exchanger.

Helium was used as a “cover gas”, to fill the space above the sodium pool over the reactor core and other areas containing sodium. Helium was chosen for the cover gas because it is very inert to chemical reactions and its insignificant activation in the presence of neutrons. Radioactive gas is stored in four shielded tanks of 2700 cubic foot

capacity at 100 psig each, until the activity level decays to a level to vent, dilute, and discharge out the building stack. Gas bottles, piping, pumps, vapor traps, valves and relief values comprise the cover gas system.

Fuel and Moderator. The SRE standard fuel slugs were slightly enriched uranium metal, 0.75 inches in diameter and 6 inches long. Fuel rods were six feet long and were formed into a cluster of seven. Rods were fitted with a helical wire to prevent contact with each other and allow the flow of sodium coolant. Hexagonal graphite blocks were used to moderate and reflect neutrons.

Fuel Handling Machine. The fuel handling machine allowed access through the plugs in the shield to the fuel for removal and placement.

The High Bay Area was the interior of the building covering the reactor.

Gas storage tanks. The gas storage tanks were connected via piping, pumps, cold traps, values, etc., to allow radioactive gases to be collected from the facility for decay to acceptable limits, and then discharged by dilution through the stack.

Building 724. This building was known as the SRE Oil Cleaning Facility. This was a peripheral building used extensively to clean contaminated sodium from pipes and miscellaneous equipment.

Section 2. **Review and Evaluation of Run 14**.

In the section entitled "Run 14", Lochbaum summarizes the events found in AI reports for Run 14 to establish his point that "evidence clearly demonstrates that the first

containment barrier between fission products produced by nuclear fuel during reactor operations and the environment had been breached for 13 of the 43 fuel elements during run 14."

The Report discusses the recorded sodium outlet temperatures, moderator temperatures, reduced sodium flows, power transients, a July 24, 1959 attempt to dislodge foreign material suspected of restricting sodium coolant flow by "jiggling" elements, the tetralin leakage problem, and the July 26, 1959 discovery of the first damaged fuel element. This section of the report acknowledges what the AI reports show by stating "Varying degree of flow blockage accounted for the temperature differences measured at the outlet of the fuel channels. Flow blockage also accounted for the power transients experienced on July 13th." Quoting directly from AI reports, this section also summarizes the post irradiation examination of the damaged fuel that lead to the fuel failure. In particular, the Lochbaum Report lists one mode of fuel failure: the formation of a low melting Fe-U eutectic caused the cladding to fail.

The Lochbaum Report does not mention a "jiggling" effort to improve coolant flow on July 20th. Also, the Lochbaum Report does not discuss the mode of fuel failure due to thermal cycling through the uranium metal α and β phase transformation temperature, which results in the fuel expanding until cladding rupture. Nor is there any mention that the fuel was bonded to the cladding with NaK.

2.1 "Jiggling". Lochbaum quotes from the AI report that "Between 0000 and 0800, on July 24, while jiggling elements in an attempt to dislodge foreign material and hence lower the fuel-channel outlet temperatures, it was noted that the elements in core channels 10,12,35, and 76 were stuck in place".[1] However, the very next sentence, not included in the Lochbaum Report, states that: "On the evening of July 22 when a similar

operation had been performed, the element in core channel 10 was free". The AI report goes on to say: "On July 26th, it was noted that the elements in channels 12 and 35 were no longer stuck. The element in channel 76 was somewhat freer than before, while the element in channel 10 was still stuck". While this information does not add much to what is already known about the SRE incident, when it is considered in the context of other available data, a somewhat different picture takes shape with respect to when the fuel failed. There is significance to the reported fact that the element in core channel 10 was determined to be free on July 22.

2.2 **Thermal Cycling.** The Lochbaum report ignores a second mode of fuel failure reported in AI report NAA-SR-4488 (suppl). Evidence of failure by thermal cycling and expansion was found in post-irradiation examination of the top section of element R-24. The AI report states: "The cladding was split…, and no evidence of Fe-U diffusion. Further evidence of thermal cycling is clearly demonstrated in Figure III-8 of the same report. The figure shows fuel temperature traces from 0800 hrs on 7-22-59 to 0800 hrs on 7-23-59 for failed element R-55. A close study of the figure shows repeated thermal cycling through the uranium metal α and β phase transformation (1220 º F) from 1200 hrs on 7-22-59 through 7-23-59 at 0800hrs. 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.[8] A third mechanism for cladding failure resulted from the NaK used to bond the fuel to the cladding.[8] Christian, pointing out that NaK boils at 1445 º F, which is 176 degrees below the boiling point of sodium (1621º F), states the fact that as the temperature exceeds 1445 º F, the vapor pressure of NaK increases exponentially. This would create large internal stresses on the cladding that could result in rupture well before the sodium in contact with the cladding began to boil.

This additional information, taken from the same AI reports referenced by Lochbaum, provides information helpful to understanding the incident. For instance, when the information is considered, the Lochbaum Report's reported failure date for all the fuel, July 13, is not so obvious. The Lochbaum Report does not consider that there is measurable and documented evidence that severe thermal cycling between July 22nd and July 23rd caused fuel rupturing. Additionally, there are the results from two recent exhaustive and detailed studies that discuss in detail the ratio of the Xe-133 to Kr-85 atom concentration found in a primary cover gas samples taken on August 2. When the omitted data is taken into account with the results of ratio of Xe-133 to Kr-85 determined in cover gas samples, it becomes very questionable that all the fuel failed on July 13, as the Lochbaum Report assumes. The experience of the element in core channel 10 does not contradict this. A detailed review of cover gas activities supports other evidence that all but one or two fuel assemblies failed around July 23.[8,9]

3.0 Radiological Consequences. In this section, the Lochbaum Report challenges statements declaring that "no radiological hazard was present to the reactor environs"[1,2] by stating that "this conclusion is not intuitively obvious given that about one third (13 of 43 fuel elements) of the SRE reactor core was severally damaged and the reactor continued to operate for nearly two weeks after this damage likely occurred (July 13, 1959)." The Lochbaum Report gives a qualitative release fraction of 15% for iodine and cesium to the atmosphere in the conclusion. To arrive at this release fraction the report establishes a lower bound for each fission product. The lower bound for cesium takes the upper bound determined by the fact that 30% of the core was damaged. For cesium, the upper bound is attenuated by assuming that 10% of the cesium was released from the fuel to the primary sodium, and 10% was released from the primary sodium to the atmosphere (.3 x .1 x .1 = 0.3). The lower bound for iodine was estimated by taking the upper bound, 30%, and attenuating that number by assuming 10% of the iodine was released from the

fuel to the primary sodium and that 100 percent of the iodine in the primary sodium was released to the environment (.3 x .1 x 1= 3). The known and reliable data for release of fission products from metal reactor fuel to the primary sodium and from the sodium cover gas is discussed below.

3.1 Release of Fission Products from Metallic Fuel. The Lochbaum Report's assumed release fraction for iodine and cesium of 10 percent is taken from a paper by Cho et. al. This report is a safety study for a sodium cooled metal-fueled reactor in conceptual design. There are no data, only assumptions for a source term for a sodium fire. Fortunately, there are data for the release of fission products from metal reactor fuel. One very important fact is that SRE was fueled with metal uranium, as opposed to uranium oxide fuels used in commercial nuclear power plants. Fission product chemistry in metal fuels is very different than fission product chemistry in oxide fuels. Experimental data and thermodynamic considerations show that fission iodine forms metal iodides in uranium metal fuels such as UI_3 and CsI, as opposed to oxide fuels where iodine can be shown to exist as I_2 and cesium forms binary compounds.[8] UI_3 and CsI salts are not volatile. Calculations show that the vapor pressures of UI_3 and CsI at the temperature of the sodium pool of SRE, 542 ºF, are 9.4×10^{-21} and 3.1×10^{-11} atmospheres respectively. The vapor pressure of I_2 at the same temperature is 66 atmospheres. To draw a comparison to a very similar but more familiar salt, the vapor pressure of table salt, NaCl, is 3.6×10^{-14} atmospheres. Additionally, experiments conducted at Argonne National Laboratory show that fission gas is not released until the fuel swells to volumes corresponding to 1 atom percent burnup. SRE fuel burnup was a factor of 10 lower or about 0.1%.[8,9] Experiments performed by this author show that iodine is quantitatively retained in the fuel at 4.2 atom percent burnup. With respect to cesium, elemental uranium atoms surround elemental cesium atoms born in the metal.

Cesium atoms contacting iodine in this environment will form CsI. Other cesium atoms will remain as elemental cesium under the strong reducing environment.

Numerous experiments have been conducted to study the release of fission products from uranium metal fuel under high-temperature conditions.[8] Results of the experiments show that fission products are not appreciably released from the metal until melting occurs. This is important because it has been shown that the metal fuel in SRE did not melt and the release of fission products resulted from uranium metal that formed the uranium-iron eutectic.[8] Release from the uranium metal forming the eutectic is also true for noble gas fission products.

In summary, fission iodine forms non-volatile salts in uranium metal fuel. Cesium atoms contacting iodine atoms will form non-volatile cesium iodide or remain in the uranium matrix as elemental cesium. The 10 percent release fraction assumed in the Lochbaum report fails to take this information into account. The release fractions reported by Daniel[9] and Christian[8] are much more reasonable with this body of knowledge taken into account.

3.2 Release from the primary coolant to the cover gas. Release fractions of 0.1 and 1.0 are used to estimate the fraction of cesium and iodine, respectively, released from the coolant to the environment. The Lochbaum Report apparently assumes that cesium is released as a volatile gas and removed by cold vapor traps and filters, and iodine is released as a gas and escapes to the environment unimpeded. The Lochbaum Report does not give any consideration to the fact that the SRE reactor core was contained in a sodium coolant system that contained 2.55×10^4 kg of sodium, and the pool of sodium directly over the core was 6 feet thick. For iodine to escape from the fuel through the sodium, it would have to pass either as elemental I_2 or sodium iodide, and make its way

from the fuel failure site up through the remainder of the core and then through 6 feet of molten sodium. First of all, iodine is formed in the fuel as a metal iodide and when released to the fuel it is released a salt, NaI. As discussed below, the vapor pressure of sodium iodide is extremely low. Even if NaI escaped from the sodium, it would quickly deposit on cooler surfaces within the reactor. If gas (I_2) was released (it was not), it would react immediately with the bond NaK, and with coolant sodium to from NaI and KI. If gas (I_2) was released in bubbles and rose to the surface, experiments show that the bubbles would collapse and elemental iodine would form NaI. If iodine escaped as a gas into the cover gas, it would react immediately with sodium vapor to form NaI. The vapor pressure of NaI at 542ºF, the temperature of the surface of the coolant sodium in SRE, is calculated to be 1.2×10^{-11} atmospheres. In other words, iodine was not released from the sodium as a gas and if it was, which it was not, it would have reacted immediately with sodium atoms in the cover gas to form NaI. The Lochbaum reported release fraction of 1.0 for iodine from the coolant to the environment did not take into account the chemistry of iodine in the fuel and the sodium and the physical properties of NaI.

The release fraction of cesium to the cover gas assumed by Lochbaum report was 0.1. Cesium, a group 1 metal, is soluble and is released into the sodium as elemental cesium or CsI. If released as CsI, the salt will react with the sodium to form elemental cesium and NaI. Using Raoult's law, and the conditions of SRE at the time the fuel damage, the fraction of cesium from 13 failed fuel elements in the cover gas can be calculated. The inventory of cesium in the failed fuel was calculated to be 0.4109 g-atoms.[8] Assuming that all of the cesium was released to sodium, the mole fraction of cesium in sodium is calculated to be 4.2×10^{-7}. Using Raoult's law and the vapor pressure for cesium at 573 ºK (573 ºF) obtains the partial pressure of cesium in the cover gas or 1×10^{-9} atmospheres. Applying the ideal gas law, one obtains the g-atoms of cesium in the cover gas to be 1.3×10^{-7} g-atoms. The resulting fraction of cesium in the cover gas is obtained

by dividing 1.3 x 10^{-7} g-atoms of cesium in the cover gas by 0.4109 g-atoms. This operation obtains a percent release fraction of 3.5 x 10^{-5}. This calculated release fraction is insignificant, and supports the conclusion that cesium did not escape from the cover gas. The Lochbaum Report fails to take any of these facts into account.

3.3 Experience at EBR-II. As an analytical chemist I am concerned with things like representative samples, valid procedures, analytical instrumentation, calibration, standards, accuracy and precision, quality assurance, etc. At EBR-II, the analytical chemistry section was responsible for analytical chemistry for the entire site. I have personally analyzed or I have had responsible for the analysis of EBR-II cover gas and coolant sodium, fuel processing, and spent fuel samples. One gains insight from these experiences. It is my experience that I-131 has not been detected in EBR-II cover gas. It is my experience that iodine has been detected in the primary sodium near the detection limit of procedures used to make the determination. The literature shows that this experience is not unique to EBR-II. This is also the experience of SRE, FERMI, and BR-5.[10]

During operation of the EBR-II, cover gas samples were taken up to three times daily during normal operations and during failed fuel events. These samples were counted on multi-channel analyzers for activity. Activities identified and quantified were X-133, X-135, Kr-85, and Ar-41. Iodine was not detected in cover gas samples during normal operations or during failed fuel events. Iodine, if present in the cover gas, would have been detected by this method. To further increase detection limits a special 8.8 liter sampler was designed to analyze cover gas. Iodine was not detected by this method.

Based on failed fuel experience, a method was developed to identify subassemblies containing failed fuel. In this technique, called “Xenon Tagging”, xenon of different

isotopic enrichments is encapsulated with the fuel and bond sodium. The concept being when the fuel failed encapsulated xenon tag gas was discharged to the cover gas along with the noble gas fission products. A representative cover gas sample was collected in a charcoal trap cooled to –78 ºC. Collection started as soon as possible after monitoring systems detected the possibility of failed fuel. The collected gas was processed to separate the noble gas fission products and tag gas from argon. The isotopic ratios were determined by mass spectroscopy. The ratios were than compared against the isotopic ratios of xenon encapsulated in fuel elements. A match of isotopic ratios identified the failed fuel. Eventually, an automated system for cover gas collection and analysis was built into a system designed to reduce, cryogenically, fission gas activity in the cover gas system. This sampling and analysis system was activated at the first evidence of failed fuel. Iodine was not detected in samples of EBR-II cover gas taken to identify failed fuel. If it were present, iodine would have been detected and measured.

It is not surprising from an analytical chemist understanding that iodine in a liquid metal cooled reactor, such as the SRE, will not migrate to the cover gas. Chemistry says that it will not be there. Sodium is a very electropositive element and iodine is a very electronegative element. Thus, these elements are extremely attracted to each other. When they do collide, they form NaI. The melting point of NaI is 661 ºC (1222ºF) and the boiling point is 1304ºC (2379ºF). Once formed in the sodium, NaI will not escape in any measurable quantities. It is extremely difficult for I_2 to exist in the environment of a sodium-cooled reactor, under the conditions experienced by SRE, EBR-II and BR-5. Physical chemists and metallurgists have studied this very closely and have published extensively on the subject. The report by Dr. Jerry Christian is an excellent review of radio-iodine behavior in metal fuels and sodium coolant including references to the literature.[8] The Lochbaum Report ignores the scientific fact that elemental iodine, if it

were released into the sodium coolant, would bond with the sodium to form NaI, preventing its release to the cover gas.

3.4 Details with respect to a "large amount of activity". Inferences that large amounts of radioactivity was discharged to the sodium coolant without talking about the quality of the fission products make the statement less meaningful. A discussion of measurable and verifiable quantitative data with respect to the chemical and physical properties of the individual fission products, the well known specific activities of individual radio-isotopes which tells something about the age of the 5,000 to 10,000 curies, the chemistry of the reactor environment, and the chemistry of metal fuels would be very helpful. It must be remembered that fission products are elements with a location clearly defined in the Periodic Table of the Elements. They have an atomic number, an electronic structure, oxidation states, a density, a diameter, etc. All of these must be taken into account when describing the environment into which they were born. In this case, a uranium metal fuel bonded to cladding with NaK. As an example, one could consider the age of the fission products. Radioactive fission products decay at a specific rate, known as their half-life. If a radioisotope has a half-life of 2 hours and there are 100 atoms, in 2 hours 50 atoms will have decayed to another element. In the case of the 5,000 to 10,000 curies reportedly discharged to the primary sodium, 80 % had a half-life of less than 1 day.[11] Half-life of some fission products and their specific activities demonstrate the point:

Radioisotope	Half-life	Specific Activities, Ci/g
Xe-133	5.243 days	1.87×10^5
Xe-135	9.1 hours	2.55×10^6
Xe-137	3.82 minutes	3.59×10^8
Xe-138	14.1 minutes	9.67×10^7
I-131	8.0207 days	1.24×10^5
I-132	2.87 hours	1.0×10^7

Kr-95	10.76 years	3.92×10^{2}
Kr-87	1.2 hours	2.83×10^{7}
Kr-88	2.84 hours	1.25×10^{7}

The Lochbaum Report does not apply this body of scientific knowledge. The only qualification provided in the report with respect to the activity estimated to be released is footnote 29 at the bottom of the page 8. With this footnote, Lochbaum states "that the reference of 5,000 to 10,000 curies does not represent or imply endorsement of this range as the true amount of fission products released to the primary sodium coolant. It merely indicates agreement that a release occurred – not the magnitude of that release". Interestingly, Lochbaum later suggests that there were far more fission products released into the SRE sodium. He lists differences in radiation readings made at Fermi vs. SRE, and the high Na-24 activity preventing the taking of a sodium sample until August 2. This will be evaluated in the next section.

3.5 Reactor Operation Details. Nine quotes from AI reports beginning on the bottom of page 5 and continuing through to the top of page 7 describe activities on July 12, 1959. The third bulleted quote summarizes events between 1530 and 2057 on the same date. The events include high bay air monitor activities at 1530 hrs and subsequent attempts to lower the activity by reducing cover gas pressure, radiation survey showing excessive activity (500 mr/hr) over a sodium level coil thimble probe located over core channel 7, activity (160,000 cpm) on a filter paper from an air monitor sample, activity and the activity decay rate for a air sample (3×10^{-7} $\mu Ci/cm^3$ after 15 min. and 4.5×10^{-8} $\mu Ci/cm^3$ after 90 min.), and a "sharp increase in stack activity (1.5 x10-4 $\mu Ci/cm^3$) at 1700 returning to normal by 2200. Activity over core channel 7 measured at 25 R/hr at 1700, decision to shut the reactor down and replace the thimble with a standard plug, initiate reduction in power at 1730, and finally by 2057 the reactor was shutdown, the drive units

removed, and the cask placed in operation. John A. Daniel, in his report "Investigation of Release From Santa Susana Sodium Reactor Experiment in July, 1959", goes into great detail to analyze the radiation and activity measurements, cover gas pressure adjustments, leak repair activities, and events surrounding the fuel handling cask ("FHC").[9] For instance, the sharp increase in the stack monitor activity coincided with the venting of the FHC during operations to replace the sodium instrument thimble with a standard plug. The fact that the high bay activity (3×10^{-7} μCi/cm^3 after 15 minutes decay and 4.5×10^{-8} μCi/cm^3 after 90 minutes of day) showed a very fast decay rate suggests short-lived noble gas fission products were present. As noted above, some of the noble gas fission products have a very short half-life. Further details describing these events can be found in the Daniel report.

At end of this section, Lochbaum concludes that the first condition necessary to release of activity to the atmosphere – damaged fuel -- was met in the SRE incident. Lochbaum then identifies the ventilation system that handled the SRE building and the system used to vent gases from the reactor. This section of the report would be more helpful and understandable if it included measurable and verifiable quantitative data with respect to the chemical and physical properties of the individual fission products, sodium chemistry, the well-known specific activities of individual radioisotopes (which would tell something about the age of the 5,000 to 10,000 curies released to the sodium), the chemistry of the reactor environment, and the chemistry of metal fuels. When all available information is taken into account with the data found in the AI reports, a reasonable timeline of known events takes shape. The Lochbaum Report section, Radiological Consequences, is very limited in its use of the available body of knowledge.

Section 4. System for Processing and Releasing Radioactive Gases. The first part of this section uses AI reports to show that radioactivity was released to the cover gas, the

shielded gas decay tanks, and to the high bay area. To document these facts, Lochbaum refers to the AI reports regarding counting rates, counts per min. (x 10^3) on filters from continuous air monitors, the history of reactor cover gas activity in $\mu Ci/cm^3$, and activities in $\mu Ci/cm^3$ in the gas decay tanks. Regarding the gas decay tanks, he says: "Unfortunately, no data was found in the documents reviewed regarding the number of (or absence of) discharges from the gaseous storage tanks following the July 13th event. But this data is available, in a November 20, 1959 inter-office report that lists dates, release rates, and the total activity for each day.[13] For the high bay area, the Report says: "Once again, it is impossible to confirm or refute the assertion that "no radiological hazard was present to the reactor environs" via the SRE building ventilation exhaust pathway. Finally, the Report claims "The scant, disconnected data prevents a quantitative assessment of the radioactivity released to the atmosphere following extensive fuel damage experienced during SRE run 14. The data show that large amounts of radioactivity released from the damaged fuel elements reached (a) the helium cover gas above the reactor pool, (b) the high bay area, and (c) the gaseous storage tanks." These statements are addressed below.

4.1 Scant and Disconnected Data. I do not agree with the Lochbaum Report's conclusion that there is scant and disconnected data regarding the release of radioactivity in the SRE incident. Since 1959, there has been extensive growth in the knowledge of sodium-cooled, metal-fueled reactors. This sense of the growth of knowledge is touched upon in the exhaustive and detailed studies of the SRE failed fuel event by Christian and Daniel. In their studies, they have included information about sodium-cooled, metal-fueled reactors available in the open literature, but that literature is not discussed at all in the Lochbaum Report. When this information is combined with their detailed review of the events surrounding the SRE fuel failure, they have uncovered ample data to support

derived release fractions not too different from those derived and reported by AI, and certainly far more reasonable than the 15% release fraction given in the Lochbaum report.

4.2 Activity in the Cover Gas. The Lochbaum Report states that large amounts of activity reached the cover gas. Questions that beg answering are: How much is a large amount and which fission products define the source of activity? The Lochbaum Report does not attempt to give an answer, but states that the activity level in the cover gas increased by a factor of "one million" between samples taken on June 20, 1959 and August 1, 1959. A study of Table IV-C-8 found in Reference 1 titled, "Activity History of the Reactor Cover Gas," show that on June 20, the activity was reported a 3.8×10^{-5} $\mu Ci/cm^3$ and on August 1 the activity of the cover gas was reported as 5.5 $\mu Ci/cm^3$. The ratio $5.5/3.8 \times 10^{-5}$ appears closer to 145,000. So whatever "large amount" means in the Lochbaum Report, it is now about a factor of seven less at least according to the table. And from information reported earlier, 80% of the activity had a half-life of less than 1 day.

Additional information is available to help understand how large is large. Daniel and Christian provide detailed information with respect their review of the literature on fission product behavior in sodium and NaK bonded metal fuels and sodium coolant. In their reports, they describe in detail the inventory of fission products at the time of the incident, the importance of fuel burnup with respect to fission product release behavior (and in particular, noble fission gas behavior), mechanisms for releasing fission products to the primary sodium and cover gas from the fuel, the venting and purging of the cover gas, cover gas samples activities, and decay corrections. They cite extensive studies in the literature, and describe their method to estimate release fractions.[8,9] Earlier in this review, the fate of iodine and cesium was discussed. Iodine and cesium does not find its way in any measurable amount to the cover gas. My experience at EBR-II shows Cs-137

was detected wherever primary sodium vapor condensed on cooler surfaces such as the rotating shield plug annulus, the stainless steel walls above the sodium pool, cold traps etc. Noble gas fission products, xenon and krypton, once released from the fuel find their way unencumbered to the cover gas, as their solubility in sodium is extremely low. It would not take much of such noble gases to obtain high readings on a survey meter such as the reading of the 2 liter gas sample (30 mr/hr).

4.3. Activity in the High Bay Area. The Lochbaum Report does not attempt to answer key issues regarding the activity observed in the high bay area. For example, what is the relative significance of the levels of activity and what were the likely fission products involved? Noble gas fission products were undoubtedly leaked to the high bay area. Daniel's reconstruction of on-going reactor operations with the time of the leaks correlate to repairing leaks discovered on July 12 at core channel 7 and on July 14 at core channels 29 and 50. It is known that fuel failed. It is known that there were leaks in the small seal plugs. It is reasonable to expect that fresh noble gas fission products would be leaked to the high bay area and be detected by continuous air monitors. It is my experience with failed fuel at EBR-II that when fresh noble gas fission products were release to the argon cover gas, noble gas leaked to the reactor environment and passed through filters of continuous air monitors. Gamma spectroscopy of filters removed from the monitors identified rubdium-88 ($t_{1/2}$ = 17.7 minutes). Rubidium-88 is the daughter of krypton-88 ($t_{1/2}$ = 2.84 hours). With such a high specific activity, it takes very little rubidium to obtain count rates on filters soaring by "20 fold" as the Lochbaum Report states. The activity and the rate of decay is consistent with fresh noble gas fission products and their daughters. This is consistent with Daniel's description of events on July 12 and July 14.

4.4 Activity in the Gas Decay Tanks. Given that noble gas fission products reached the gas decay tanks, the questions needing a response are how much is "large" and what are

the identities of the fission products? It is clear that noble gas fission products were intentionally transferred to the gas decay tanks. Release fractions determined by Daniel and Christian, when applied to the SRE failed fuel core inventory of xenon and krypton, represented a small fraction of the krypton and xenon inventory in the SRE fuel. But this small amount of activity would increase the activity in the gas decay tank by many orders of magnitude, as shown in Table IV-C-10 in reference 1 and pointed out by Lochbaum.

4.5 Radiation at stack release point and number, timing, and release of gas from gaseous storage tanks. The Lochbaum Report expresses concern that no data was found on either the radiation levels at the stack release point or on the number, timing, and radioactivity level of release from the gas storage tanks. AI report NAA-SR-4488 gives stack monitor affluent activity on July 12 at 1700 to be 1.5×10^{-4} $\mu Ci/cm^3$ and on July 15 at 0600 hours to be 7×10^{-5} $\mu Ci/cm^3$. The report goes on to say that the activity level remained intermittently high for about 5 hours. In addition, a November 20, 1959 inter-office report gives the quarterly tabulation of radioactive gases released to the atmosphere from SRE decay hold up tanks from July 1, 1959 to October 1, 1959. The Report lists dates, release rates, and the total activity for each day. The gap noted between 7/25/59 to 8/22/59 to 9/16/59 is explained as being due to slow release rate.[13]

4.6 Data from inside ventilation duct work. Activity found in the upstream duct work vs. activity down stream is addressed in the Lochbaum report. The table reproduced in the Lochbaum Report is activity in $dpm/100cm^2$, taken from ductwork in Building 724.[12] This is a peripheral building called the SRE Oil Cleaning facility. This facility was used extensively to remove contaminated sodium from pipes and miscellaneous equipment. Since the data is for a peripheral building, a connection between the SRE ventilation system and Building 724 seems doubtful. Lochbaum qualifies the data as "indirect", suggesting that the filters removed activity but not all of the activity. Without knowing

from the Lochbaum Report anything about the building, the significance, if any, of the data with respect to the release between July 12 and July 26, 1959 is questionable.

4.7 Comparison of SRE with Fermi. A comparison between the SRE reactor and the Fermi is discussed in the Lochbaum Report. The differences in radiation readings in primary loop piping and at the immediate heat exchanger are noted in order to hypothesize that the amounts of fission product activity released into the primary sodium system was significantly higher than the amount released into the Fermi sodium. Activities reported to show higher radiation readings were taken from G.B. Zwetzig, Atomics International, "Survey of Fission and Corrosion Product Activity in Sodium or NaK Cooled Reactors," AI-AEC-MEMO-12790, February 28, 1969. The report discusses possible reasons for the different readings such as power runs, cold trap operation, etc. All are very reasonable. On close examination the 11R/hr radiation reading is for a deposit found in the SRE intermediate heat exchanger. A point source reading is something totally different than a general radiation field. This comparison of SRE with Fermi is anecdotal, and does not provide the necessary data to support much higher release of fission products as suggested in the Lochbaum Report.

4.8 The August 2 Sodium Sample. The validity of the August 2 sodium sample is questioned in the Lochbaum Report. The challenge is supported by 1) a suggestion, with no data, that quantities of activity discharged to the primary coolant may have been significantly higher overwhelming the "gettering" action of tetralin and tetralin decomposition products, and 2) reproducing a quote from a AI report that says, "The Na^{24} activity in the primary sodium coolant delayed the procurement of the first sodium sample until August 2, about 7 days after the reactor shutdown. It should be noted that during this interval, circulation and cold-trapping of the primary sodium continued; thus

the loss of some portion of the fission product activity originally present prior to the first sample was certainly possible".[3]

A review of the referenced AI report does give measured data to evaluate Lochbaum's claim. Table V of report NAA-SR-6890 shows a comparison of fission product activities measured in the Aug 2 sample with fission product activities measured in the Oct. 31 sample. Activities for the Aug 2 sample are decayed to Oct. 31 for the comparison. It is a fair assumption that the primary sodium continued to be circulated during this time period to remove residual and decay heat from the reactor. The ratio of the predicted Oct 31 to measured Oct 31 show that Sr^{90}, Sr^{89}, Ce^{141}, Ce^{144}, Zr-Nb^{95}, and Ru^{103} are much less than predicted. From my experience I would suggest this is in fact plate-out as suggested in the AI report. The data for cesium and iodine show that they are more in line with what was expected even if the differences between the two samples for cesium and iodine is 64% and 37%, respectively. To an analytical chemist this is not surprising. There is a very high probability that these differences are the result of sampling and analysis.

For example, experience at the analytical laboratory at the EBR-II site sheds light. Bulk liquid sodium from the primary coolant is sampled at typical reactor coolant temperatures. Liquid sodium is drained into a container, allowed to cool, and transferred to the laboratory in a inert gas to protect the integrity of the sample. Initially, there were difficulties performing cesium determinations until it was discovered that when sodium cools below its melting point, cesium migrates to the cooler surfaces of the container and the surface of the sodium. Not understanding this obtained low and non-reproducible data. Information on cesium migration may not have been available to analytical chemists in 1959, since this discovery was not made at the EBR-II analytical laboratory until the late 1960's. With respect to iodine, Table V of Reference 3 shows I-131 activity to be 0.00012μCi/g sodium and the predicted activity to be 0.00019μCi/g sodium. This is

not unexpected. Considering that the first sample contained a lot of activity, 0.42 μCi/g sodium and the second sample had only 0.00012 μCi/g sodium, a difference of 37 percent can be expected. Procedures used in those days involved the dissolution of the sodium sample to prevent loss of iodine, conversion of iodide to iodate than to I_2. The iodine was extracted into carbon tetrachloride to separate it from other fission products, back extracted into a aqueous solution, and the iodine precipitated as either silver or palladium iodide. The precipitate was collected on a filter paper, dried, and counted. The counter registered counts per minute. Counts per minute were correct for geometry, branching ratios, etc. to obtain disintegrations per second (d/s). Disintegrations per minute were then converted back to μCi. Recoveries for such methods could range for 60 to 90 %. From memory, I would place an error on this method at around 15 to 20 percent. When all is considered with respect to the sodium sample, the Lochbaum challenge can be answered with measurable data, and is shown to have little substance.

5. Review and Evaluation of References. The Lochbaum Report lists references to suggest reasons why the reported inability to detect iodine in the cover gas at SRE and Fermi as unexplained and inconsistent with numerous experimental finds. Comments about the references follow.

K. Haga, Y. Nishizawa, T. Watanabe, S. Miyahara, and Y. Himeno, "Equilibrium and Nonequilibrium Partition Coefficents of Volatile Fission Products Between Liquid Sodium and the Gas Phase," Nuclear Technology, Vol. 97, No. 2, pp. 177-185, February 1992. Estimates of NaI(g) based on the Haga literature using SRE data at the time of the July 13 excursion results in 0.002 Ci of I-131 in the He as NaI(g) using Castleman's work referenced in the Haga paper. Using Haga's extrapolated approach, one obtains 0.0003 Ci of I-131 in the He as NaI(g). Iodine-131, if it was in the gas as NaI, would condense on the cooler walls of the cover gas region. This report is not contrary to the body of

knowledge showing the iodine is not detected in the cover gas of sodium cooled metal fuel reactors.

Suguru Tashiro and Horihiko, Ibaraki University, "Diffusion Coefficient Determination of Sodium Iodide Vapor in Rare Gases with the Use of Ionization Sensor," Journal of Nuclear Science and Technology, Vol. 38, No. 7, pp. 551-556, July 2001. This paper describes a method to determine diffusion coefficients by a method combining the analysis of measured diffusing mass with continuous monitoring of the sodium vapor concentration in flowing stream of sodium iodide-rare gas mixtures. It does not describe the release of NaI from damaged fuel to the primary sodium and than to the cover gas. In fact, sodium is not part of the development of the method. The conditions under which the sensor was tested have nothing to do with the conditions of the SRE. The body of scientific data, reactor operating experience, sodium chemistry, iodine chemistry, metal fuel chemistry shows that iodine is not discharge to the cover gas. This paper is not an appropriate reference. Also, see response to Haga et.al. above.

Ahmed Hasan, Yasser T. Mohamed, and Tarek F. Mohammaden, New Mexico Technological University, "Waste Generated from LMR-AMTEC Reactor Concept, Waste Management Conference, Tuscon, Arizona, February 23, 2003. This paper presented at the Waste Management Conference is concerned with defining waste generation and characterization for the LMR-AMTEC, and identifying applicable U.S. regulations the govern waste transportation, treatment, storage, and final disposition. It is not a technical paper describing the fate of iodine in a sodium-cooled metal-fueled reactor.

Young Ho Cho, Mee Jang, Young Wook Lee, Moon Soo Park, Chang Sun Kang, Chang Hyun Chung, Do Hee, Hahn, and Joo Hyun Moon, "The Analysis of Radiological

Consequences Due to Hypothetical Core Disruptive Accident in KALIMER," Proceedings of the ICAPP "03. This paper describes a safety study for the Korean Advanced Liquid Metal Reactor (KALIMER). The containment performance of the KALIMER was analyzed for the Hypothetical Core Disruptive Accident with the postulated fission product release with the relevant requirements. The conceptual design for KALIMER is under development in Korea. For the accident in question, sodium pool fires and spray fires have been assumed to follow the HCDA as design basis events to identify whether the containment could maintain integrity. This paper is not appropriate for the SRE event.

None of the references offered support the Lochbaum claim "that the reported inability to detect radioiodine in the cover gas of SRE and Fermi-I is unexplained and inconsistent with numerous experimental finds".

Conclusion

The qualitative assessment given in the Lochbaum Report that it seems reasonable that – "say 15 percent" – is a reasonable release fraction for Cs-137 and I-131 from the SRE to the atmosphere is without technical merit. The Lochbaum Report quotes selectively from AI reports in an anecdotal way to arrive at the qualitative release fraction. This qualitative release fraction is not supportable by the years of liquid metal cooled reactor operating experience, research into the behavior of fission products in metal fuels and sodium coolant, and the laws of chemistry and physics. It is contrary to my experience as an analytical chemist at a liquid metal fast breeder reactor, EBR-II.

References

1. NAA-SR-4488, “SRE Fuel Element Damage, Interim Report,” Atomics International, Nov. 15, 1959.

2. NAA-SR-4488(suppl), “SRE Fuel Element Damage Final Report,” , Atomics International, 1961.

3. NAA-SR-6890, “Distribution of Fission Product Contamination in the SRE,” R.S. Hart, Atomics International, 1962.

4. NAA-SR-5898, Metallurgical Aspects of SRE Fuel Element Damage Episode,” October, 15, 1961.

5. NAA-SR-6359, “SRE Core Recovery Program,” December 1, 1961.

6. AI-AEC-MEMO-12790, “Survey of Fission Products and Corrosion Product Activity in Sodium and NaK Cooled Reactors,” February, 28, 1969.

7. ESG-DOE-13403, “Sodium Reactor Experiment Decommissioning Final Report,” Aug 15, 1983.

8. Chemical Behavior of Iodine-131 during SRE Fuel Element Damage in July 1959, Jerry D. Christian, May, 26, 2005.

9. Investigation of Releases from Santa Susana Sodium Reactor Experiment in July, 1959, John A. Daniel, Sr., Daniel & Associates, Inc., May 27, 2005.

10. A.W. Castleman, , Jr, “LMFBR Safety, I. Fission Product Behavior in Sodium”, Nuclear Safety, Vol. 11, No. 5, Sept.-Oct. 1970.

11. Robert W. Dickinson, Nucleonics, Vol. 18, No. 1, January, 1960.

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13. G. Borg, “Quarterly report of activity released to atmosphere,” Nov. 20, 1959 inter-office letter to W.L. Fisher.