

Flow Cells for Energy Storage

Workshop Summary Report

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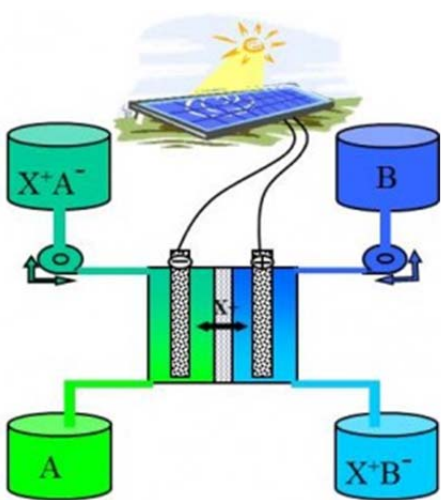
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U.S. DEPARTMENT OF
ENERGY

Energy Efficiency & Renewable Energy *Coordinated with:* Office of Electricity Delivery & Energy Reliability



Executive Summary

An essentially identical technology to a reversible fuel cell is that of a redox flow cell (RFC) or redox flow battery (RFB), where a RFC can be seen as merging the concepts of RFBs with recent improvements in fuel cells. To investigate how a RFC can be a grid-scale electrical-energy-storage (EES) system and the associated technological needs, this workshop was held. The specific objectives of the workshop were to understand the needs for applied research in RFCs; identify the grand challenges and prioritize R&D needs; and gather input for future development of roadmaps and technical targets for RFCs for various applications.

In the context of an electricity grid with rapidly changing needs due to expanding variable renewable generation, RFCs offer promise for providing many essential grid-scale services. RFCs possess inherent technical attributes that make them well suited to meet a wide range of different EES needs, and they have the potential to be the chosen technology to serve much of this growing EES demand. The basic technology is proven, but not (yet) cost effective. However, some recent breakthroughs in RFC technology, such as order-of-magnitude improvements in cell power density should enable RFC systems with significantly lower costs. Therefore, the future potential of RFCs is promising and activity in industry is also on the rise.

However, there has been very limited RFC technology development over the past three decades, and the technology is ideally suited for fuel-cell and battery developers to make game-changing improvements. What is required is a development path that is uniquely grounded in scientific advances, where breakthrough after breakthrough emerges from the close collaboration of scientists and technologists. This is similar to the approach and execution of fuel-cell research and development by the Department of Energy (DOE) Energy Efficiency and Renewable Energy (EERE) Fuel Cell Technologies Program (FCTP). For the various EES applications, technical targets and roadmaps need to be developed similar to the multiyear Research Development and Demonstration (MYRD&D) plan of FCTP, which contains specific targets in terms of the dominant phenomena (e.g., durability, performance, crossover, etc.) and components (e.g., catalyst, membranes, stack, etc.). In addition, the Energy Storage Program within the DOE Office of Electricity Delivery and Energy Reliability (OEDER) has shown and should continue to demonstrate their excellent progress in field demonstrations (*i.e.*, $TRL \geq 6$) of a variety of EES devices utilizing their good working relationships with electric utilities and other potential EES customers. ARPA-E has also already demonstrated how their unique DOE mission can have a large impact on RFC technology, and funding of truly breakthrough RFC-related concepts should continue to be supported by ARPA-E, but a growing program in applied research, development, and demonstration to examine many of the common and synergistic RFC components and phenomena is still required, which is outside the scope of ARPA-E's mission.

Besides the grand-challenge need for a well-managed, comprehensive program on RFCs, specific technological hurdles are identified in order to reduce RFC system cost. Traditional chemistries can be utilized but there is a need for greater understanding and optimization of the intrinsic processes that are occurring including catalysis and transport phenomena. Durability and lifetime as well need to be improved and are some of the key limitations of the technology. Similar to fuel cells, individual components and diagnostic methods need to be studied and improved. Finally, there is also a recognized need to investigate some new RFC chemistries including solid suspensions and non-aqueous ones, although this requires fundamental research.

Table of Contents

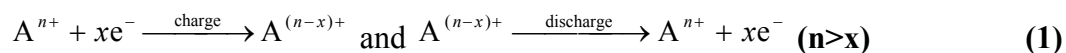
Executive Summary	ii
Table of Contents	iii
Introduction.....	1
Background.....	3
Need for Electric Energy Storage (EES)	3
Issues for adoption of EES.....	9
EES Technologies and the Need for RFCs.....	10
RFC Research	14
Historical.....	14
Current Research, Development, and Demonstration.....	15
Comparing RFCs and Fuel Cells	18
Breakout Sessions	21
Breakout I.....	21
A. Metal/flow systems	21
B. Novel and nonaqueous systems.....	24
C. Traditional single and multiphase systems.....	25
Breakout II	28
A. Catalysts/electrode designs	28
B. Membranes	30
C. Stack, system, and related components.....	31
Literature Cited	33
Appendix A: Workshop Agenda.....	35
Appendix B – GRIDS Program Targets	36

Introduction

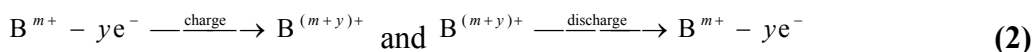
Renewable-energy sources, such as solar and wind, are being deployed in larger numbers than ever before, but these sources are variable and often unpredictable. Analysis suggests that an electric grid could become destabilized if non-dispatchable renewable energy exceeds 20 percent of the energy-generation capacity without energy storage.¹ If electricity is to be stored, it must first be converted to some other form of energy. There are some technologies that enable practical storage of energy at their current levels of deployment, but only a very small fraction of North American power plants employ such technology (about 2% of the installed generation capacity).^{2,3} To ensure that renewable energy succeeds in delivering reliable power to US consumers, the nation needs cost-effective and reliable storage at the grid scale. The need for energy storage has been identified as being sufficiently significant that it is specifically called out for consideration in the Energy Independence and Security Act of 2007.⁴

Hydrogen and other chemicals are considered to be potential energy storage options to enable increasing the renewable energy content of the electrical grid. Nominally, this storage is derived through reversible or regenerative fuel cells, those that operate both in the discharging and charging states. A workshop was held on H₂/O₂ reversible or regenerative fuel cells in April of 2009.⁵ It was determined that reversible fuel-cell technology is feasible for cost effective storage of renewable electricity, with further development required. Specific recommendations included the development on new catalysis materials for air/oxygen electrode; increased durability for both low- and high-temperature systems; new stack designs and new approaches to heat management; and increasing the speed or ramp rate for the systems so that they can be response to changes in the generation and load demands.

A redox flow cell (RFC) or redox flow battery (RFB) is essentially an identical technology to a reversible fuel cell, where the connotation of RFC suggests the leveraging and knowledge transfer of fuel cells to traditional RFBs. Such a device allows one to remove the catalysis constraints by examining different electrochemical couples that are more efficient than H₂/O₂. While such systems are expected to have lower gravimetric and volumetric power and energy densities than reversible fuel cells, this is not a primary concern for grid-scale energy storage where efficiency is much more favored over footprint. Although the electrochemical couple is different for RFCs, many of the components are the same as are the flowing of reactants in a fluid phase coupled with catalytic reactions in the electrodes. RFC half reactions can be written as



and



for the anode (negative electrode) and cathode (positive electrode), respectively.

These reactions occur in a power module as shown in Figure 1. Since the reactive components can be stored in tanks outside of the electrochemical stack, energy and power requirements can

be sized separately, as in a fuel cell. This is much different than in containerized batteries where both energy and power must be scaled together, resulting in substantially higher costs when larger quantities of energy are required but not necessarily more power and this also leads to much less flexibility for future load/design changes.⁶ In addition, storage can be increased with relative ease and minimal cost compared to the stack, which is often the most expensive component; and as such, RFCs are very attractive for longer duration and larger energy and power applications (i.e., grid-scale electrical energy storage (EES)). Because of the decoupling of energy and power in RFC configurations, both cost per unit of power-generation/storage capability (\$/kW) and the cost per unit of energy-storage capacity (\$/kWh) can be considered. As renewable or stationary power-backups, RFCs can provide rapid startup, load follow prices or peaks, and generate high-quality electricity under safe and economical conditions.

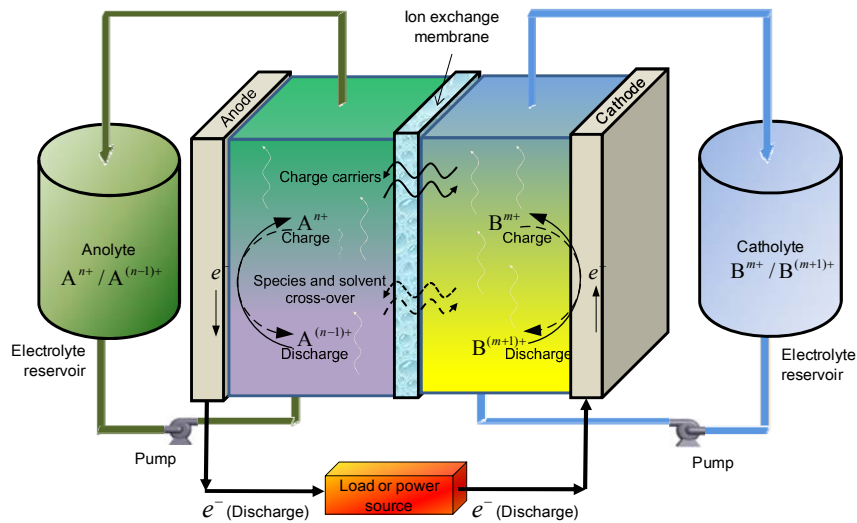


Figure 1 – Schematic of a RFC system.

This workshop was held to investigate the appropriateness of RFCs as grid-scale EES systems and what the associated technological needs are. The overall purpose of the workshop was to understand the applied research and development needs and the grand challenges for the use of RFCs as energy-storage devices. Specifically, the objectives were to understand the needs for applied research in RFCs; identify the grand challenges and prioritize R&D needs; and gather input for future development of roadmaps and technical targets for RFCs for various applications. The workshop focused namely on applied R&D needs (TRLs between 1 and 6), and on the general RFC needs and not specific chemistries or couples.

Background

Joe Eto, Imre Gyuk, and Mike Perry discussed the needs for electric energy storage (EES) and the favorable attributes of RFCs in meeting them.

Need for Electric Energy Storage (EES)

The current worldwide electric generation average power was estimated to be about 20 trillion kilowatt-hours per year (~2.3 GW) in 2007.⁷ More than two-thirds of the current mix is from some form of fossil fuels, with most of the balance coming from nuclear and hydroelectric power generation. At present, only about 3% comes from renewable-energy technologies, although the annual new energy-storage deployment is expected to grow from 121 MW in 2011 to 2,353 MW in 2021.⁸ Furthermore, developing economies and electrification of the transportation sector both point to strong year-over-year growth in terms of electrical demand.

The present electric grid constitutes an enormous physical infrastructure, with a near-instantaneous transmission of value from primary power sources and generation assets to end users. Utilities, regional transmission organizations (RTOs) and independent system operators (ISOs) contract with customers who can allow interruption of power to their operations in exchange for compensation. If a system operator encounters an unexpected need for reserve power, they may temporarily disconnect these contracted customers to restore reserve availability until demand falls or additional generation comes online. Striking this balance between demand and supply could, of course, be provided by electrochemical energy storage, charging the storage asset during periods of excess supply and drawing from the storage asset when supply is interrupted or when the system experiences higher-than-anticipated demand. The need to handle the difficult-to-predict aspects of consumer demand has resulted in the construction of power plants that may only operate for 100 hours a year or less. Generators are dispatched to respond to small oscillations in demand over very short time scales. Generators are also turned on and sped up to meet increasing load during the peak time of the day. Storage is a vital tool that would uncouple customer demand from the generation side of the grid—allowing vital flexibility in control and maintenance of the electric grid.

There is an increasingly recognized need for energy storage on the grid scale, which derives from the fact that there is a substantial projected growth of renewable energy generation both domestically and globally, as witnessed in Figure 2, and these are variable generators that are readily affected by the local weather conditions. In addition, this projected growth is a reflection of the strong desire to increase the mix of renewable energy generation. For example, at least 29 states have adopted renewable-energy portfolio standards (RPS) with ranges of 10 to 40% of the generation to come from renewables (e.g., California has an RPS of 33% by 2020). While almost all of the current renewable generation is in the form of hydropower (see Figure 2), growth is severely limited as it requires certain geological formations and waterways.

The perceived technologies for increased renewable generation are namely wind and solar power, of which wind is the most promising and in development in the short to medium time horizon. However, as noted above, both of these are variable. For example, a solar PV farm can have an 80% change in 5 minutes due to clouds passing over the array. Another example is given in Figure 3, which shows the hourly wind output from the Tehachapi Wind Farm, which demonstrates considerable variability both throughout the day as well as day to day variations.

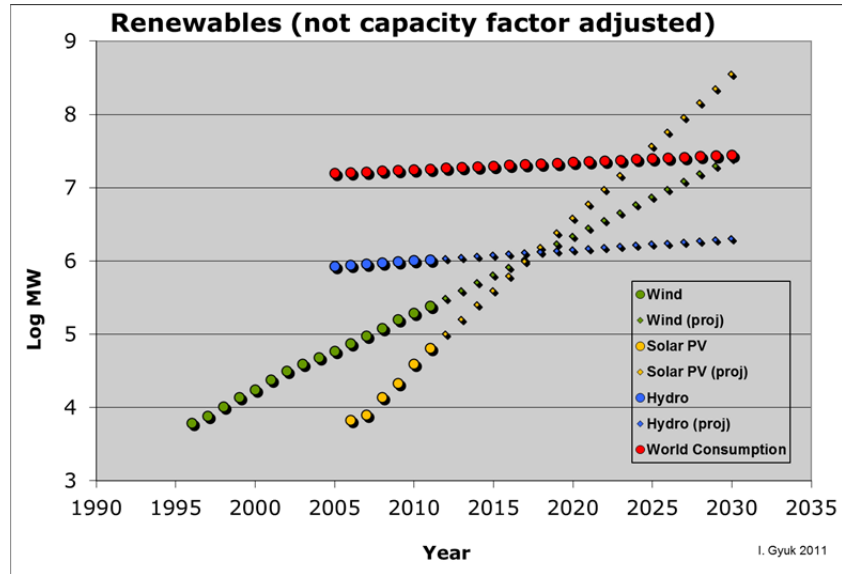


Figure 2 – Actual and projected growth of renewable energy generation and consumption worldwide.

In fact, it is clear that on some days the wind did not blow and the generation was zero, even demonstrating a net negative load on the grid. Although the monthly average is relatively stable, the hourly and daily fluctuations can represent an issue for use of the wind energy as a part of the baseload grid generation. This issue is highlighted in Figure 4, where one readily sees how the net load varies due both to the variable load, which is more or less predictable, and the wind output, which is not.⁹ These variations are currently smoothed by less efficient coal- and natural-gas fired peaker plant generators or spinning reserves due to the lack of EES. Because of its variability and the mismatch between peak demand and peak wind power output, there is significant value added by the incorporation of storage.¹⁰

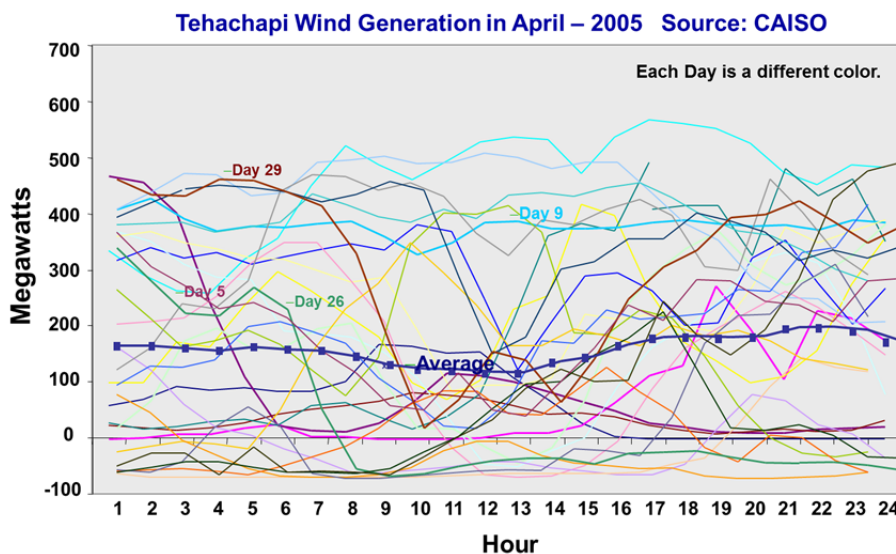


Figure 3 – Tehachapi wind generation by hour for each day in a month.

To understand the various markets and application for EES, different studies have been accomplished, with varying results due to different underlying assumptions.¹¹⁻¹³ An example of such a report is that from Sandia National Laboratory which conducted a study for EES related to grid-

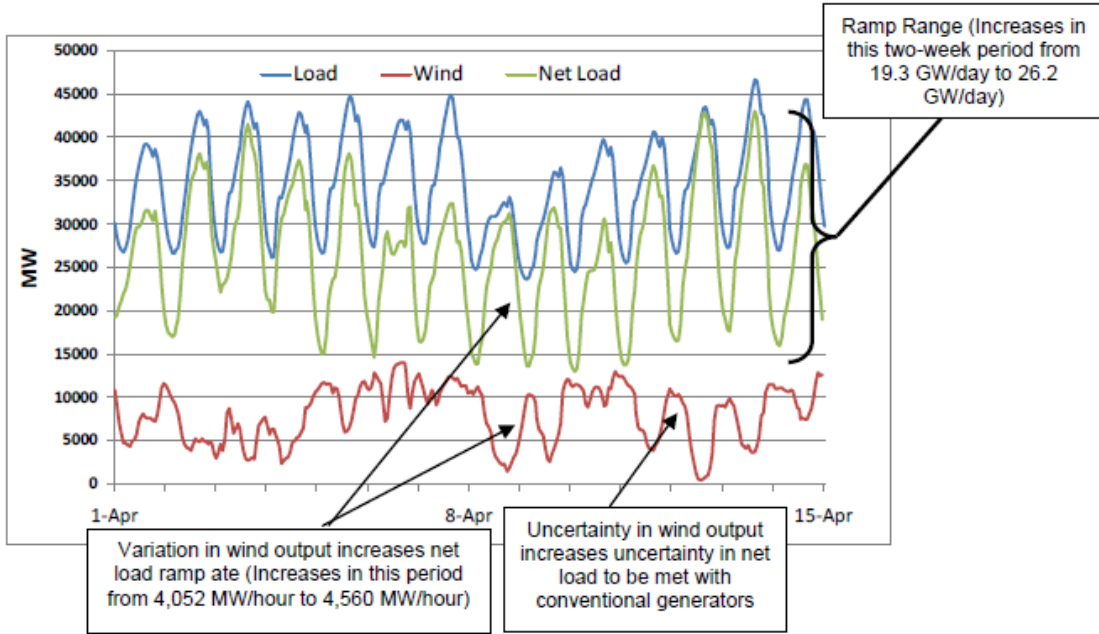


Figure 4 – Data for two weeks showing the wind generation, grid load, and the net grid load.

scale activities.¹¹ The main areas can be separated in terms of their storage time and power requirements as shown in Figure 5. From Figure 5, one can see that the storage needs span a large range in both metrics, thus suggesting that a portfolio of technologies would be best suited for adoption; however, a single technology that can meet multiple demands would be very desirable and impactful. To quantify the impact and opportunity for the different applications in Figure 5, the Sandia report provides some high-level estimates of the economic impact and the value proposition for each application as shown in Table 1. In the table, the key value propositions and high-impact applications are circled and discussed in more detail below. These applications are essentially, renewable firming or the integration of renewables to the grid but in a stable, constant fashion; arbitrage or load following over the day (minutes to hours response times) to account for the tariff and time-of-day pricing structure; and frequency regulation which is high value but low overall

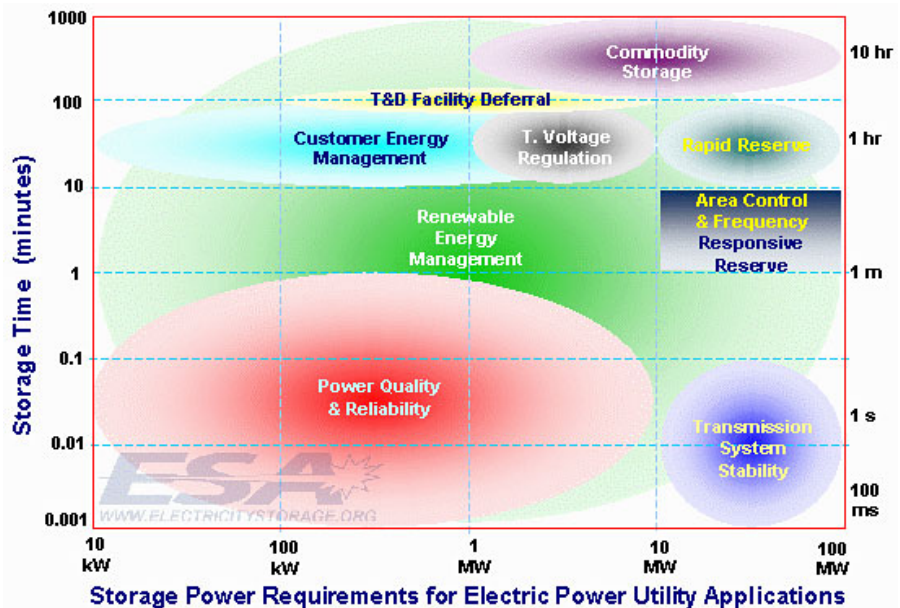


Figure 5 – Different storage applications and their expected time and power requirements from the Electricity Storage Agency.

economic impact and requires response on the order of seconds and may represent a possible early market adoption scenario for grid-storage technologies.

Table 1 – Summary of the different applications for grid-scale EES including the necessary metrics, possible economic and power impact and cost value.¹¹

#	Benefit Type	Discharge Duration*		Capacity (Power: kW, MW)		Benefit (\$/kW)**		Potential (MW, 10 Years)		Economy (\$Million) [†]	
		Low	High	Low	High	Low	High	CA	U.S.	CA	U.S.
1	Electric Energy Time-shift	2	8	1 MW	500 MW	400	700	1,445	18,417	795	10,129
2	Electric Supply Capacity	4	6	1 MW	500 MW	359	710	1,445	18,417	772	9,838
3	Load Following	2	4	1 MW	500 MW	600	1,000	2,889	36,834	2,312	29,467
4	Area Regulation	15 min.	30 min.	1 MW	40 MW	785	2,010	80	1,012	112	1,415
5	Electric Supply Reserve Capacity	1	2	1 MW	500 MW	57	225	636	5,986	90	844
6	Voltage Support	15 min.	1	1 MW	10 MW	400		722	9,209	433	5,525
7	Transmission Support	2 sec.	5 sec.	10 MW	100 MW	192		1,084	13,813	208	2,646
8	Transmission Congestion Relief	3	6	1 MW	100 MW	31	141	2,889	36,834	248	3,168
9.1	T&D Upgrade Deferral 50th percentile ^{††}	3	6	250 kW	5 MW	481	687	386	4,986	226	2,912
9.2	T&D Upgrade Deferral 90th percentile ^{††}	3	6	250 kW	2 MW	759	1,079	77	997	71	916
10	Substation On-site Power	8	16	1.5 kW	5 kW	1,800	3,000	20	250	47	600
11	Time-of-use Energy Cost Management	4	6	1 kW	1 MW	1,226		5,038	64,228	6,177	78,743
12	Demand Charge Management	5	11	50 kW	10 MW	582		2,519	32,111	1,466	18,695
13	Electric Service Reliability	5 min.	1	0.2 kW	10 MW	359	978	722	9,209	483	6,154
14	Electric Service Power Quality	10 sec.	1 min.	0.2 kW	10 MW	359	978	722	9,209	483	6,154
15	Renewables Energy Time-shift	3	5	1 kW	500 MW	233	389	2,889	36,834	899	11,455
16	Renewables Capacity Firming	2	4	1 kW	500 MW	709	915	2,889	36,834	2,346	29,909
17.1	Wind Generation Grid Integration, Short Duration	10 sec.	15 min.	0.2 kW	500 MW	500	1,000	181	2,302	135	1,727
17.2	Wind Generation Grid Integration, Long Duration	1	6	0.2 kW	500 MW	100	782	1,445	18,417	637	8,122

*Hours unless indicated otherwise. min. = minutes. sec. = seconds.

**Lifecycle, 10 years, 2.5% escalation, 10.0% discount rate.

[†]Based on potential (MW, 10 years) times average of low and high benefit (\$/kW).

^{††} Benefit for one year. However, storage could be used at more than one location at different times for similar benefits.

Non-dispatchable, variable generation assets (such as solar and wind) require fast-response grid-reliability services. Also referred to as ancillary services, these inject or withdraw real power on the grid to adjust and stabilize grid power quickly. When the supply of power does not match actual load on a moment-by-moment basis, the imbalance will cause the grid frequency to deviate from the standard 60 Hertz. Minor frequency deviations can damage electric devices. Major deviations and imbalances cause generation and transmission equipment to separate from the grid, and in the worst case can lead to cascading blackouts. Grid fluctuations can occur on several timescales: sub-second, second, minute, hour, day and season. The most important of these timescales for grid stability are the sub-second, second, and minute imbalances which cause frequency fluctuations. Today, marginal frequency regulation capacity is provided by natural gas combustion turbines. Using fossil-fuel combustion turbines to support grid reliability

at short time scales increases the wear, fuel consumption, and emissions of these generation assets, decreasing their lifetime and potentially causing regulatory issues related to environmental permitting.¹⁴ In addition, relying upon fossil generation resources leads to inefficiencies in frequency correction. Large-scale gas turbines change output at longer timescales than desired for grid reliability, forcing grid operators to commit additional regulation resources to compensate.¹⁴

Providing grid-reliability services necessitates specific performance characteristics including:

- Low costs: the incumbent natural gas turbines establish the cost benchmark.
- High-power capabilities: the more power a resource can provide, the greater the market opportunities.
- High ramp rate: frequency deviations occur on the sub-second and second scale.
- Accurate response: higher accuracy will be more important as new market tariffs which pay premiums (or deduct penalties) based upon the accuracy of a resource's response come into fruition.
- Long cycle life: regulation resources may be called upon for either ramp-up or ramp-down services thousands of times in a year, especially since a fast-ramping high-value storage device will rarely be at rest for long periods.

EES can also provide renewable integration or renewable firming where the output of the wind farm is essentially modulated such that the utility does not see the strong variations (see Figure 3). Thus, renewables integration can mean a range of services that improve the economic value of the energy generated by renewable generation assets. This range of services includes the grid reliability mentioned above, as well as time shifting or arbitrage, where one stores the energy generated during low-value periods and delivers it when at higher or peak periods where it is the most valuable. Wind energy is a good candidate as it typically generates most of its electricity at night (e.g., see Figure 3) where it has lower value. In fact, it is expected that regulation and load following will increase from anywhere between 10 to 200% with more wind scenarios among the different ISO areas.¹⁵

As one moves from the wind generator to a utility standpoint, EES has advantages in possible maximization of utilization of existing generation and transmission and distribution (T&D) resources. Thus, EES can be placed at the site of choke points in the distribution network or at a substation to improve local power quality and reliability.¹⁶ Placing storage at these locations also allows for deferral of some capital improvements; thus making optimization of an improved T&D system considerably easier. For example, a T&D substation may be nearing its capacity limit, indicating a need to upgrade its capacity in the near future. However, it may be nearing its capacity only for infrequent, peak loads, which could possibly be covered by an EES system. While this is not a large market, T&D deferral is attractive for the high value placed on electricity delivered for this purpose.¹¹

As one moves from the utility to the customer or behind-the-meter, EES can be used for retail time-of-use arbitrage, which is essentially a time-shifting application at a customer site to take advantage of peak versus off-peak electricity pricing. In addition, EES can be utilized to also ensure uninterruptable power supply for data centers, hospitals, and other customers with

critical-load requirements. Such an application does not depend on renewable integration to benefit from EES. Of course, a key metric in this application is the difference between the peak and off-peak prices as it will determine the minimum roundtrip EES system efficiency. One of the major concerns is the reliability of the storage since if it fails the whole economic gain can be wiped out due to a high peak cost depending on the tariff structure at the customer location.

Table 2 - Key performance targets for grid-storage applications, from Reference ¹⁷

Application	Purpose	Key Performance Targets
Area and Frequency Regulation (Short Duration)	Reconciles momentary differences between supply and demand within a given area	Service Cost: \$20 per MW Roundtrip Efficiency: 85%–90% System Lifetime: 10 years Discharge Duration: 15 minutes–2 hours Response Time: milliseconds
Renewables Grid Integration (Short Duration)	Offsets fluctuations of short-duration variation of renewables generation output Accommodates renewables generation at times of high grid congestion	Roundtrip Efficiency: 90% Cycle Life: 10 years Capacity: 1–20 MW Response Time: 1–2 seconds
Transmission and Distribution Upgrade Deferral (Long Duration)	Delays or avoids the need to upgrade transmission and/or distribution infrastructure Reduces loading on existing equipment to extend equipment life	Cost: \$500 per kWh Discharge Duration: 2–4 hours Capacity: 1–100 MW Reliability: 99.9% System Life: 10 years
Load Following (Long Duration)	Changes power output in response to the changing balance between energy supply and demand Operates at partial load (i.e., increased output) without compromising performance or increasing emissions	Capital Cost: \$1,500/kW or \$500/kWh Operations and Maintenance Cost: \$500/kWh Discharge Duration: 2–6 hours
Electric Energy Time Shift (Long Duration)	Stores inexpensive energy during low demand periods and discharges the energy during times of high demand (often referred to as arbitrage)	Capital Cost: \$1,500/kW or \$500/kWh Operations and Maintenance Cost: \$250- \$500/kWh Discharge Duration: 2–6 hours Efficiency: 70%–80% Response Time: 5–30 minutes

The different EES applications have different acceptable costs and total power and duration requirements. As such, it is difficult to target a single metric that can concisely address the ultimate cost target for grid-based EES. As one moves from the utility to the customer or behind-the-meter, EES can be used for retail time-of-use arbitrage, which is essentially a time-shifting application at a customer site to take advantage of peak versus off-peak electricity pricing. In addition, EES can be utilized to also ensure uninterruptable power supply for data centers, hospitals, and other customers with critical-load requirements. Such an application does not depend on renewable integration to benefit from EES. Of course, a key metric in this application is the difference between the peak and off-peak prices as it will determine the

minimum roundtrip EES system efficiency. One of the major concerns is the reliability of the storage since if it fails the whole economic gain can be wiped out due to a high peak cost depending on the tariff structure at the customer location.

Table 2, as taken from a report prepared by the Nexight Group based upon a workshop convened by Sandia, PNNL, and the Minerals, Metals, and Materials Society (TMS) for the US Department of Energy, suggests various cost and performance targets for key utility applications.¹⁷ The key metrics for an EES are the response time, discharge time, power capacity, roundtrip energy efficiency, calendar and cycle lifetimes, and operating and capital costs. It is also key to understand the needs and who the customer is in terms of ISO, utility, generator, or end user. However, as can be readily seen in Table 2, not all of this relatively short list of key metrics has been defined for even this small number of “high-impact” utility applications. Furthermore, it should be noted that the targets presented in Table 2 do not agree with those presented in Table 1. For example, with respect to Capital Costs in Table 1, it does not make sense that a customer would pay more for an EES product than the 10-year Lifecycle Benefit for the application shown in Table 2. Clearly, there is a pressing need to quantify accurately the key market requirements for at least for some EES applications.

Issues for adoption of EES

Although a compelling case can be made for EES, especially going forward, there are some issues that are inhibiting the adoption of EES. First, the markets for renewables integration are not yet mature in that there are regulatory and perception issues. However, these are slowly being overcome by education and demonstration projects. Furthermore, it is believed that there will be economic incentives in the future for renewable generation assets to integrate EES and provide steady, high-quality power to the utilities instead of the current must-buy agreements.

There are also significant regulatory issues with other, non-renewable applications. For example, for T&D deferral, only a few storage devices have been approved and classified as transmission assets. It is only recently that the traditional grid reliability services rules have started changing as ISO/RTOs respond to FERC Order 890, promulgated in February 2007, which requires grid operators to modify tariffs and rules to allow non-generating resources, such as EES and demand-response technologies, to participate fully in the regulation services market. In response to FERC’s orders, ISO/RTOs have started to allow regulation to provide compensation for beneficial energy withdrawals in addition to energy injections. Thus, although slow to change, it does seem as if the regulations are becoming more open to the concept and idea of EES systems. For example, although the idea of a storage portfolio standard is very new and many questions about its logic and utility remain to be answered, California has taken the initial steps to determining its feasibility in Assembly Bill 2514, which directs study towards energy storage implications and possibilities.

However, these various regulatory issues and the immaturity of renewables are not the primary reasons why distributed EES technologies have not yet been widely adopted or commercialized. The primary barrier to widespread adoption is that existing EES solutions have not been cost effective, especially considering the competition of generator sets and spinning reserves. If EES products could provide a good economic benefit, then they would have been adopted despite these regulatory issues (*e.g.*, in behind-the-meter applications, where utility regulations are

irrelevant) and the regulatory issues would have been addressed previously if there were actually legitimate economic incentives to do so. First and foremost, EES must demonstrate both low capital and maintenance costs (i.e., long lifetimes and high durability); this is where the key technical R&D challenges arise.

EES Technologies and the Need for RFCs

The above discussion highlights some of the various applications for EES systems. However, due to their wide possible operating range (see Figure 5), there are various technologies that can be pursued. One can thus compare the performance of different EES technologies to the metrics of storage time and power requirements as shown in Figure 6. For example, for power quality, such as frequency-regulation applications, one can see that fly wheels and supercapacitors have the appropriate storage time and power attributes. For energy management and longer time storage, one sees that different battery chemistries are possibilities, with the target zone for RFCs noted in orange. Of course, a technology that can meet multiple requirements is of more value (e.g., a long-term storage technology that also has a high ramp-rate capability).

One can also compare the technologies based on their cost points for the most impactful applications (see Table 1) as shown in Figure 7. In this metric, RFCs or flow batteries are shown being able to only meet a few of the most impactful markets. Current estimates of costs for conventional batteries and flow batteries are significantly higher than the required targets: a 2008 estimate of RFB costs suggested nearly \$2500/kW, albeit without specification of duration or sizing.¹⁸ A reduction in cost, especially per unit power, of an order of magnitude (or even greater than 2X) would open up most of the markets and applications to RFCs or various other batteries. Regardless of detail, however, significant cost reduction must be achieved. Technological improvements, material development, and economies-of-scale must be achieved to ensure success in the marketplace. Of course, one must again be cognizant that the main competition for EES systems in terms of costs (assuming no carbon-emission costs) are gas turbines that are ubiquitous, well understood, and widely accepted. Combined-cycle turbines however are typically only 60% fuel-to-electricity efficient with spinning reserves on the order of 35 to 40% efficient with ramp rates of zero to full power in 10 minutes.¹⁹

To compare the different technologies on a generation basis, one can look at the levelized cost of electricity (LCOE). This has a few key components that factor into the calculation: up-front capital cost;

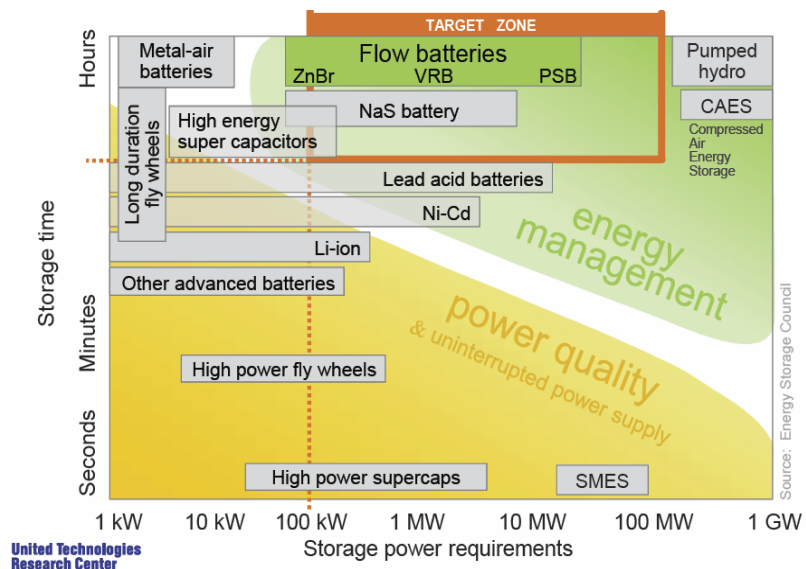


Figure 6 – Storage time and power for various EES systems.

operations and maintenance cost; capacity factor; lifetime; and ongoing variable fuel costs. While the LCOE for a natural gas turbine depends heavily on its capacity factor, most are typically in the range of \$280/MWh. For hydropower and compressed air energy storage (CAES), LCOEs are around \$100/MWh assuming long lifetimes (50 years).⁹ However, while both of these technologies are inexpensive and possess high energy-storage capacities, they require the appropriate geologic formations which are not necessarily optimal located near renewable generation sites. For battery systems, the LCOE is dependent mainly on capital cost and roundtrip efficiency, thus one wants to minimize total system cost while maintaining high roundtrip efficiency, something that requires an integrated device rather than a separate fuel cell and electrolyzer.

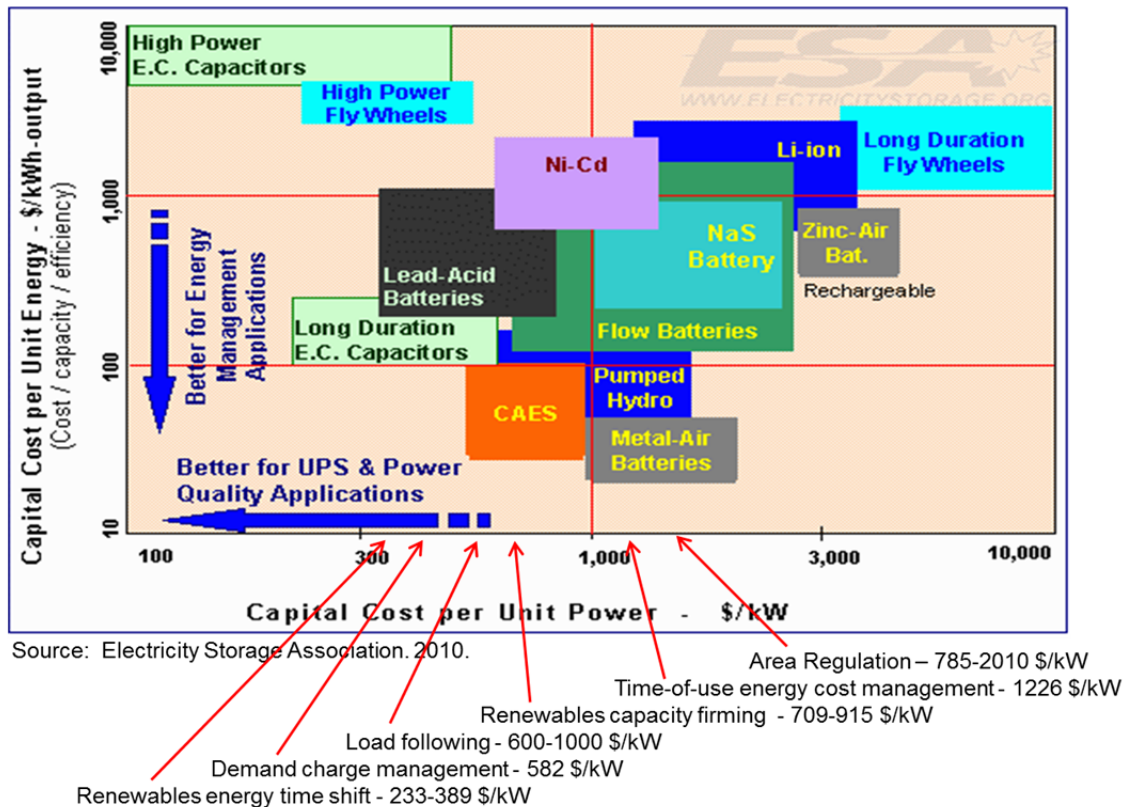


Figure 7 – Cost per energy versus cost per power for different technologies with the most impactful applications and their associated costs from Table 1 shown.

Upon comparing alternate EES technologies to CAES and hydropower, and assuming greater than 5000 cycles with an 80% roundtrip efficiency, ARPA-E initially determined the metrics given in Appendix B for its GRIDS (Grid-scale Rampable Intermittent Dispatchable Storage) FOA. While these metrics are a good first step in terms of developing technical targets, a more in-depth study and roadmap is warranted. It should also be emphasized, that as the GRIDS Program name suggests, the focus of this set of technical targets was for smoothing of intermittent sources (*i.e.*, renewables), and these are not all necessarily applicable to other EES applications (e.g., the minimum operating time at rated power, or discharge duration is ≥ 2 h for many of the applications shown in Table 1).

For the various proposed electrochemical EES technologies, one can examine the merits of them. Figure 6 and Figure 7 demonstrate that most containerized batteries seem to lack either the necessary energy storage capacity or power, especially in terms of cost for long-term storage. Of the containerized batteries, the most promising are sodium sulfur, lead acid, and Li ion. NaS batteries are constrained to a 6-hour discharge and are currently sold in 1MW, 6MWh units by NGK Insulators, the company that developed the battery with Tokyo Electric Power Company. They currently cost about \$3200/kW (\$530/kWh) and have recently had some safety concerns. Lead-acid batteries may be the most technologically mature batteries installed on the grid today. They suffer from severely limited cycle lifetimes of 2200-4500 cycles, 5 to 7 year calendar lifetimes and associated higher operating and maintenance costs, but their low installed cost (around \$1000/kW) and proven reliability make them a common choice for current wind co-location projects.²⁰ Li-ion batteries have shown promise although they are still costly (\$1,000-4,000 \$/kW) and there are safety concerns with these as well, especially for large systems as required for EES applications.

In general, containerized batteries have fundamental issues that make them ill-suited for large-scale EES. These issues are namely that power and energy are not independent, which limits modular flexibility and also results in expensive replacement costs. In addition, they have relatively low active-material-to-inactive-material ratios (typically $\sim \frac{1}{2}$ cost, weight, & volume) which results in poor scale-up costs. They also have relatively short cycle life with deep charge/discharge cycles due to electrodes physiochemical changes during cycling. Safety is inherently challenging since the reactants cannot be easily isolated. In comparison, RFCs do not suffer from these issues due to their ability to separate energy and power capacity and costs, thus minimizing total system cost while maintaining high roundtrip efficiency. Table 3 shows a qualitative comparison between these two different types of battery architectures.

Table 3 – Comparison between conventional or containerized and flow batteries.

Key Attribute	Conventional	Flow
Energy Density	+	-
System Complexity	+	-
Inherent Safety	-	+
Deep Cycle Life	-	+
Cell-to-cell Uniformity	-	+
Power / Energy Independence	-	+
Capital Cost* (\$/kWh)	Inactive materials scale with Energy (kWh)	Inactive materials scale with Power (kW)

* Active material cost depends on chemistry; however, active-material utilization will depend on architecture (e.g., depth of discharge (DoD) limits, accessibility)

To understand the interplay between costs and performance for the different systems and the power of the energy/power separation provided by RFCs, one can look at a figure of merit like in Figure 8. One can see that since the main costs for RFCs is in the stack or power unit, the costs

come down appreciable with power density, which is one area of active research where both UTRC and LBNL have demonstrated power densities in excess of 1300 mW/cm^2 for vanadium and H_2/Br_2 chemistries, respectively; until recently, the best reported power densities for most types of RFCs was around 150 mW/cm^2 . Furthermore, compared to a conventional containerized battery, RFCs can tolerate higher initial stack material costs since for longer discharge times, they will effectively become less expensive due to the fact that they scale with power and not energy (i.e., $\$/\text{kW}$, not $\$/\text{kWh}$). This is because only more reactants are required to add more energy capacity to a RFC, whereas in a conventional containerized battery, one must add additional cells that contain both the inactive cell materials, as well as additional reactants.

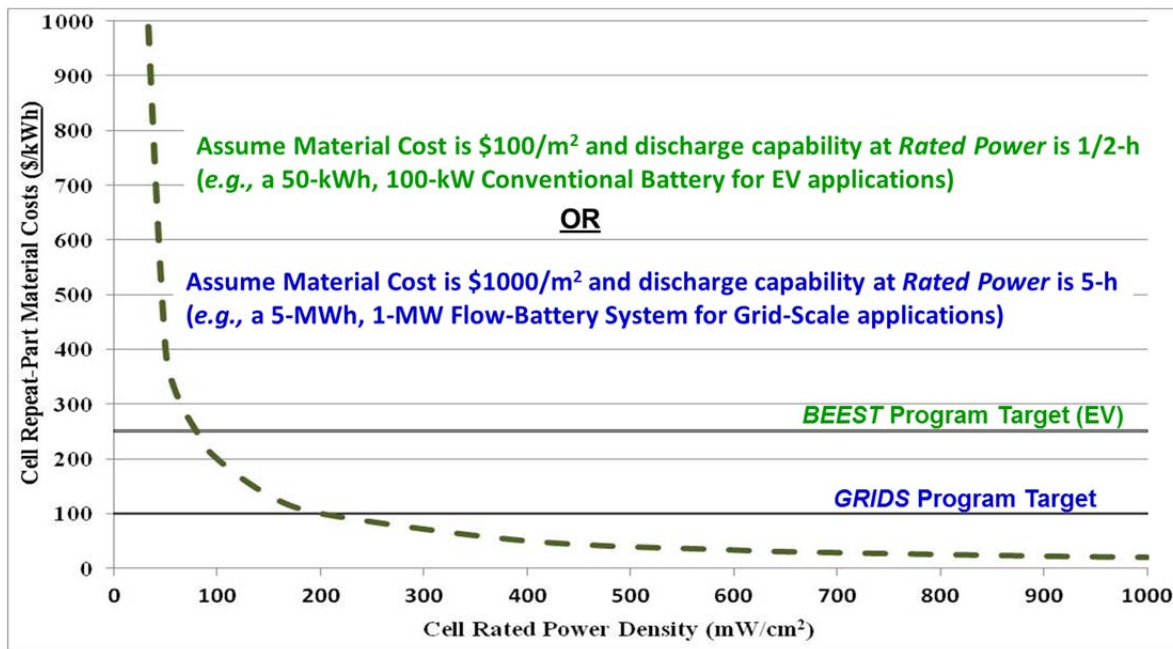


Figure 8 – Cell repeat costs as a function of cell power density for a containerized batter and RFC and also shown are the ARPA-E targets for the BEEST and GRIDS programs.

In the context of an electricity grid with rapidly changing needs due to expanding variable renewable generation, RFCs offer promise for providing essential grid services. Their flexibility allows them to meet a range of different needs, and they have the potential to be the chosen technology to serve much of this new demand. RFCs enable:

- High energy capacities and high power-density cells
- Minimal non-active material costs
- Independently sized power and energy and thus are easily scalable and upgradable
- High utilization of active materials
- Inherently safer storage of reactants
- Good round-trip energy efficiencies
- Long cycle life

The basic technology is proven, but not (yet) cost effective. The future potential of RFCs is promising. There has been very limited development over the past three decades and the technology is ideally suited for fuel-cell and battery developers to make game-changing improvements, such as the recent breakthroughs in power density that have been realized.

RFC Research

RFCs have been researched for many decades, although active mainly in the 70's and 80's and recently in the 2010's. There are several excellent reviews of RFCs/RFBs.²¹⁻²⁶ Below, some historical perspectives provided by Bob Savinell and Steve Clarke are given, followed by current research funded by DOE under the Fuel Cell Technologies Program (FCTP), Fuel Cells Research and Development subprogram (Dimitrios Papageorgopoulos), Office of Electricity Delivery & Energy Reliability (OEDER) (Imre Gyuk), and ARPA-E (Mark Johnson).

Historical

Redox-flow-based energy-storage technologies, which RFCs belong to, were originally developed in earnest in the 1970s for NASA,^{23,21} although the earliest RFC concept is thought to be a $\text{Br}_2/\text{Sn}^{2+}$ system proposed by Posner.²⁷ NASA originally examined the Fe/Ti system where the cost estimates were \$190 to \$330 per kW in 1975. NASA also screened redox couples including Fe²/3 on C, Cr²/3 on C and on B₄C, V²/3/4/5 on C and B₄C, etc. and chose Cr²/3. They also screened a substantial library of possible metal and metalloid catalyst materials. These cells had 2 to 10 ohm-cm² resistance for commercial/in-house anion exchange membranes (vs. 0.2 ohm-cm² for Nafion 117). Cr²/3 wasn't 100% efficient in charging due to some H₂ generation. They added a fuel cell to extract energy from the H₂ produced.

The structure of the cell was plate/frame structures similar to current fuel cell stack designs. They noticed significant shunt currents, where in a 40 cell stack they saw a ~2% loss at 65 mA/cm². Shunt current losses increase with the square of the number of cells, and while one can decrease the shunt currents by increasing inlet length, this causes increased pumping losses. NASA tried a Fe/Cr mixed reactant solution, which allowed them to use a lower resistance cation-exchange membrane instead of anion-exchange membranes.

For stationary, grid-scale applications, the requirements and metrics are quite different than those of NASA, and the technological progress on them has been impeded by the lack of understanding of the underlying transport and reaction mechanisms in the RFC geometry. For example, most RFCs have low power densities, equivalent to where low-temperature fuel cells were decades ago.²⁸ Today, the knowledge gained in these related fields can be leveraged and a renaissance in RFCs initiated, such that the performance, durability, and cost targets for a large-scale energy storage platform realized.

In addition to the traditional couples above, there are non-aqueous systems like Zn/Ce, which AIC invented in 1999. The chemistry operates in methyl sulfonic acid (MSA) and provides good performance, yet the company floundered due to inadequate resources although several stack design improvements were made. The non-aqueous RFBs have the theoretical ability to obtain higher energy densities than the aqueous systems due to their larger voltage-stability window; however, this increase is tempered by the move to more expensive, toxic, and dangerous solvents. This is especially true when one realizes a key benefit of a RFC is to have the cost centered around the stack, which can be readily improved upon or minimized by increasing the amount of reactant (*i.e.*, increased energy storage). What is required for commercialization of a

RFC is high intrinsic reversibility of the electrode reactions and easy transport through engineering design resulting in high-power-density stacks, long lifetimes and cycle life, and inherently safe and inexpensive reactants.

Current Research, Development, and Demonstration

As noted above, RFC research has been recently rediscovered due to advancements in fuel-cell technology as well as the renewed interest in renewable energy generation and the subsequent need for grid-level EES. For optimal resource allocation, it is suggested that these RFC projects at the respective DOE offices leverage off of each other, with each one taking ownership of projects that are within their established sphere of expertise.

Within FCT, Versa Power is working on R&D related to a reversible solid-oxide fuel cell such that it can operate efficiently both as an electrolyzer and fuel cell. The key is operating at higher temperatures, which allows one to overcome some of the inefficiency provided by the oxygen reactions. They have demonstrated less than $0.3 \text{ } \Omega\text{-cm}^2$ resistance and an operating lifetime of over 1000 hours at 500 mA/cm^2 .

Within ARPA-E, transformative projects are being analyzed through the GRIDS (Grid-scale Rampable Intermittent Dispatchable Storage) program, where the effort is to bring game-changing ideas and concepts from the laboratory across the funding “valley of death” to the marketplace. These projects exist in industry, academia, and national laboratories. One issue however is that there are also some seedling projects that hold great promise but require more substantial long-term R&D, something that a DOE R&D office would be ideally suited to take over.

In terms of RFCs and semi-RFCs (i.e., one electrode is a plating metal), there are several projects that are being investigated. Examples include a flow-assisted rechargeable Zn-MnO₂ battery by CUNY Energy Institute. The concept is to use non-toxic and inexpensive, and water-compatible chemicals. Existing performance has shown over 500 cycles in 1.5 Ah batteries, and scale up is commencing. General Atomics is working on using MSA and new RFC designs to make a better lead-acid battery that overcomes the cycle-life issues mentioned above.

The University of Southern California is working on metal-air semi-RFC by utilizing iron, which is inexpensive and readily available. The low-cost battery is expected to have a very long lifetime. Primus Power is working on a system concept wherein they have long lifetime (~20 year) 200 kW contained cells using a metal-halogen semi-RFC. Similarly, Case Western Reserve University (separately, non ARPA-E funded) is working on an all-iron RFC where iron ions are used instead of air.

Similar to the Versa Power reversible fuel cell is that of Proton OnSite, who is working with a reversible polymer-electrolyte fuel cell and not a solid-oxide one. The concept is to use alkaline conditions and membranes in order to reduce the catalyst costs. They are leveraging their experience with electrolyzers and hydrogen compression.

United Technologies Research Center and partners are examining using fuel-cell designs and understanding with the vanadium system and have already shown an order-of-magnitude increase in the power density of these cells. Such increase allows for the cost benefits seen in Figure 8 to begin to be realized.

Similarly to the UTRC design, Lawrence Berkeley National Laboratory and partners are working with H₂/Br₂ RFC, where they have used fuel-cell engineering and designs to demonstrate high power densities (1.3 W/cm²). The system is shown to be very reversible with minimal side reactions due to the lower operating voltage of the couple.

Within OE, work is focused both on R&D and, more heavily on, later stage deployment and demonstration of technologies. They have successfully shown how they can leverage their money to get industrial and venture capital support and cost share to demonstrate and deploy technologies at scale. The R&D efforts are centered at Sandia National Laboratory and Pacific Northwest National Laboratory where work is on finding new RFC couples as well as developing them into stacks and prototype systems.

Under \$185M in American Recovery and Reinvestment Act (ARRA) funding, OE initiated many storage demonstration projects for a variety of storage applications including RFCs. These RFC-based projects are:

- Primus Power: 25 MW / 3 hour battery plant for firming 50 MW of wind, replacing \$75M of gas-fired generation. Zn-halogen electrolyte
- Premium Power: 5 x 500 kW (2.8 MWh energy capacity) ZnBr batteries that are truck-mobile
- Ashlawn battery for Painesville, OH: vanadium redox batteries (1 MW, 8 MWh) for bulk storage / peak shaving at a 32 MW coal power plant.
- Enervault: 250 kW / 4 hour Fe-Cr flow battery for 300 kW of PV

These demonstrations are clearly showing that aggregated storage of distributed generation can behave differently than individual generation and requires different management strategies.

There have also been some companies such as VRB Power Systems and Plurion that have tried to commercialize the technology. For example, multiple fielded RFC demonstrations have been done, especially with VRB. For example, Sumitomo Electric Industries (SEI) has installed a number of relatively large VRB systems in Japan, as shown in Table 4.

Many of these demonstrations have been modestly successful except for the system cost. An example of a fielded prototype unit (VRB Power Systems) was a 500-kW / 2-MWh plant in Moab, Utah, which supplied T&D upgrade deferral in an environmentally-sensitive site. It started in 2004 and ran for 5 years with 1600 cycles completed.

Finally, it should be noted that there are also some RFC projects funded by NSF and DOD. Even with the above projects and current popularity, funding of RFC R&D is sparse and often poorly applied. Many researchers are walking down the same blind alleys that others have walked down before and there needs to be a concerted, sustained effort with a systematic, target approach for effective RD&D to be accomplished and the technology realized.

Table 4 – Fielded demonstrations of VRB Systems by Sumitomo Electric Industries.

Location Type	Intended Application	System Ratings (Power / Energy)	Initial Operation
Office Building	Peak Shaving	100-kW / 800-kWh	Feb 2000
Utility Company	Peak Shaving	200-kW / 1600-kWh	Jun 2000
Research Institute	Wind Turbine Stabilization	170-kW / 1000-kWh	Mar 2001
Golf Course	PV hybrid	30-kW / 240-kWh	Apr 2001
Factory	Peak Shaving / UPS	1500-kW / 1500-kWh	Apr 2001
Utility Company	Peak Shaving	250-kW / 500-kWh	May 2001
University	Peak Shaving	500-kW / 5,000-kWh	Jul 2001
Office Building	Peak Shaving / UPS	120-kW / 960-kWh	Jun 2003
Factory	Peak Shaving / UPS	300-kW / 1200-kWh	Aug 2003

Comparing RFCs and Fuel Cells

Along with Mike Perry and Steve Clarke, Tom Zawodzinski discussed the synergies between fuel cells and RFCs and what can easily be transferred or not. In many ways, the current state of technology of RFCs is similar to the state of fuel cells about one to two decades ago.

Essentially, a discharging RFC is a fuel cell. Thus, RFCs are different than fuel cells in that one has to consider charging as well as discharging behavior. The charging behavior can be quite different and is often more challenging than discharging due to perhaps irreversible reaction mechanisms and additional degradation due to the higher potentials. For example, materials that are robust in discharge reactions often fail under charging (*e.g.*, carbon). In addition, good discharge catalysts may be poor charging catalysts.

Similarly, one also needs to be cognizant of the relevant battery metrics and the dependence of performance on state-of-charge (SOC), something fuel cells do not concern themselves with. The transport and chemistry are much more complex in the electrodes and membranes in RFCs compared to fuel cells, but this also gives more handles to explore the system behavior. This complexity arises in that there are many different RFC electrochemical couples and often can be in very disparate phases from slurries to aqueous and nonaqueous solvents to gases. Typically, liquid is chosen due to its easier ability to store the reactants, although these must be able to be stored in the charge state for extensive periods of time. Furthermore, the couples are typically in supporting electrolyte or at least involve many different ion speciation. The many different chemistries also mean that impurities and standards, which are important, remain unknown for most RFC technologies.

Unlike traditional fuel cells, RFCs are typically highly efficient and thus are dominated more by ohmic and mass-transfer losses rather than kinetic ones. In addition, compared to fuel cells, RFCs have different cost drivers and energy density and power density concerns, making cost modeling much more complex but also with perhaps a higher payoff. An RFB must power itself and this imposes complex tradeoffs, for example:

- High voltage couples and reactive chemistries vs. materials of construction
- Low voltage couples and low reactive chemistries vs. large footprint and electrolyte lakes
- Shunt (bypass) current losses vs. pumping losses
- Expensive catalysts vs. electrolyzer power density
- Good reaction kinetics and temperature vs. materials

Although the chemistry and controlling phenomena may be different in RFCs compared to fuel cells, there are also many synergies and commonalities, where the advances made in the past decade in terms of fuel-cell components and diagnostics can be leveraged to great advantage for RFCs. As shown in Figure 1, the main components of a RFC are the same as in a fuel cell, namely current-collector plates, anode, cathode, and separator. Thus, advances in next generation catalysts and catalyst structures, bipolar plates, manufacturing, etc. can all be

translated to RFCs. Table 5 gives a component by component comparison between fuel cells and RFCs.

Table 5 – Comparison of typical fuel-cell versus flow-cell (battery) components and key challenges.

COMPONENT	HYDROGEN FUEL CELL	FLOW BATTERY
REACTANTS	GAS	MOSTLY LIQUIDS, UNDERSTANDING SPECIATION
ELECTRODES	Pt/C THIN LAYER, DURABILITY, SUBSTRATE STABILITY	GRAPHITE THICK LAYER FELT, CATALYST, DURABILITY, SUBSTRATE STABILITY
ELECTRODE DESIGN	THIN LAYER MEA Low C (concentration), high D (diffusivity)	FLOW THROUGH High C, low D
MEMBRANE	CATION PERMSELECTIVE IN GAS PHASE, LOW REACT SOLUBILITY	CATION, ANION, OR MICROPOROUS IN LIQUIDS, HIGH REACT SOLUBILITY
BIPOLAR PLATES	GRAPHITE, METALS, COMPOSITES	SAME
CELL STRUCTURE	PLATE AND FRAMES WITH FLOW FIELDS	SAME, LIQUID HYDROSTATIC EFFECTS, SHUNT CURRENT PATHS
BALANCE OF PLANT	H ₂ STORAGE, WATER BALANCE, THERMAL CONTROL, POWER CONDITIONING	TANKS, PUMPS, POWER CONDITIONING, C/D control

From the table it is clear that there are both similarities and differences between the systems. Furthermore, even if the same component can be used in both systems, unexpected and complex interactions can occur. For an example of the complexity, Figure 9 shows how a typical perfluorinated sulfonic acid (PFSA) membrane conductivity changes when immersed in sulfuric acid. Surprisingly, the conductivity mainly decreases with increasing acid concentration, showing perhaps an effect of dehydration and ionic condensation within the membrane's conductive pathways. Such multiple components make the transport much more complex for the RFC, even if a porous separator and not a membrane is used.

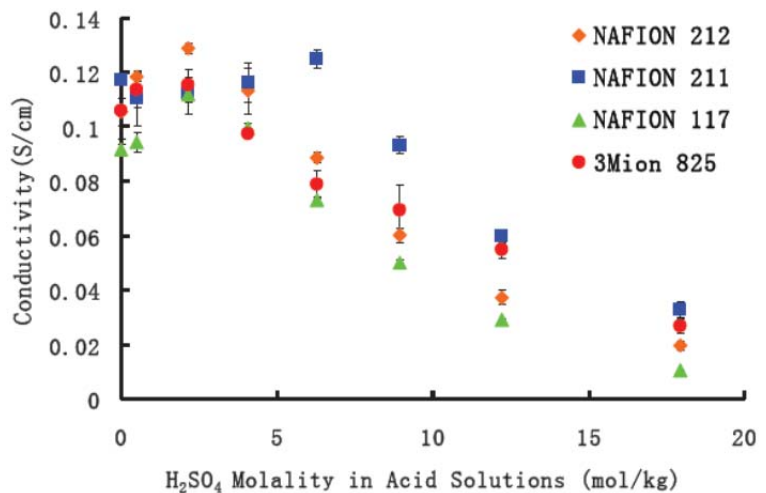


Figure 9 – PFSA membrane conductivity as a function of sulfuric acid molality.

Perhaps one of the greatest benefits that can be translated from fuel-cell to RFC research is the development of testing protocols for various

metrics including lifetime, performance, etc. and associated advanced in-situ and ex-situ diagnostics. For example, one can adapt the thin-film rotating disk electrode techniques to study kinetics on relevant materials, except using carbon fibers instead of Pt/C particles. In addition, one can also utilize the segmented cells developed for understanding heterogeneities among the planform, something that may be more significant in RFCs, especially as they may relate to shunt currents.

RFCs are a lot like fuel cells and hence one can study them as such by utilizing impedance data, polarization curves, and steady-state operation. These approaches are typically not the way battery scientists test the system and can provide new insights into the controlling phenomena and underlying physics by examining the different regions of operation. In addition, as RFCs are batteries, many of the standard battery diagnostics (cycling, depth-of-discharge and peak-power pulses, effect of C-rate on capacity and performance, etc.) can also be utilized and the requisite Ragone, Peukert, and other identifying plots developed. As both battery and fuel-cell diagnostics can be leveraged, this holistic approach can provide for relatively quick advancements in the technology.

While many fuel-cell diagnostics can be utilized by RFC, some of them are not transferrable due to having a liquid electrolyte and the nature of the electrodes which become more flow-through rather than flow-by. Examples are hydrogen or CO adsorption/stripping voltammetry to determine electrochemically active surface area, detailed understandings of the flow and current distributions including what goes where and how fast, and life testing and durability methods which are more system dependent.

Although RFCs are much more complex chemically than fuel cells, many of the advancements in mathematical modeling can be transferred. For example, the various multiscale modeling approaches are ideal for studying the complicated, typically non-ideal solution properties and accounting for speciation and such effects as those that lead to Figure 9. The models can be developed, calibrated, and validated by comparison with experimental data. In fact, due to their complexity, modeling is much more critical and impactful in terms of describing the behavior of a RFC than for a fuel cell.

Breakout Sessions

Two sets of three breakout sessions each were undertaken. The specific instructions to the breakout sessions were to choose a spokesperson or scribe (typically the facilitator) and to discuss and prioritize the R&D needs for the specific given topic as they relate to RFCs. The different sessions and subtopic for the breakout sessions were:

- Breakout I: Phenomena
 - Focus on performance, power, durability, lifetime
 - Metal/flow systems
 - Novel and nonaqueous systems
 - Traditional single and multiphase systems
- Breakout II: Components
 - Focus on diagnostics, materials, manufacturing, and modeling needs
 - Catalysts/electrode design
 - Membranes
 - Stack, system, and related components

Breakout I

Individuals wrote down ideas on 3x5 cards, these were then each discussed and sorted to identify commonalities. From there, they identified those that were particularly important for RFCs and prioritized the research needs. Further, each idea was categorized based on what issue it addressed: durability, cost, or performance.

A. Metal/flow systems – facilitator Thomas Fuller

The group first discussed what constituted metal/air flow batteries. The consensus definition was a RFC where one of the electrodes was a metal, and therefore deposition and stripping was required. This is also commonly described as a hybrid RFC. Although many of the challenges with metal/RFC are common to other RFCs, the group focused on those that were unique to the metal/flow systems.

Uniform deposition and stripping of metals (durability): Since the metal/RFC implies that metal is being plated and stripped during charge and discharge, this technology has all of the challenges of any secondary battery that has a metal electrode. The issues with using metal lithium in lithium-ion cells are well known. Dendrites form easily, particularly at high charging rates. Dendrites can reach the other electrode and short the cell—they can also break off and thus reduce the useable metal. The advantages are understood, but the durability of existing systems is low. Understanding the formation dendrites and identification of methods to mitigate their formation are needed.

Development of highly loaded electrodes with equal performance (cost): If the active material is contained in the electrode, then one of the key advantages of the RFC system is lost—namely the decoupling of power and energy. Typical porous electrode design emphasizes either power or energy. The likely applications for RFCs require high energy, which for the metal cell may require low power. As the power per unit electrode area or active material is decreased, the size of the cells will increase. Since cost is largely driven by size, the lower the loading the higher the cost of the battery. Research is needed to understand the trade-off between power and energy for the metal electrode design as well as novel approaches to achieve highly loaded electrodes with good performance.

Creation of stable colloidal dispersions of metal that can be charged/discharged (cost): An alternative to the highly loaded electrode is the use of a colloidal dispersion of active material that can be charged and discharged. This concept would maintain the key advantages of RFCs – decoupling of power and energy. However, this concept is not yet well developed. Research is needed to identify novel active materials, to develop dispersions, and conceive feasible system designs.

Tools to characterize electrodes (performance and durability): Compared to the fuel-cell and traditional battery technology, tools to characterize catalysts, electrodes, and cells are not well developed for RFCs. Although many are not directly applicable, much of the understanding from the fuel-cell and battery programs can be leveraged. Of highest priority is knowledge building, with a secondary priority on the establishment of standards and metrics. Considering the diversity of technologies being pursued, it is too early to set rigid protocols, and targets other than high level system metrics. The recommendation is for evolving guidelines. That being said, there is tremendous need to develop robust and discriminating tools to evaluate materials and design both for initial performance and life. These tools would include electrochemical performance, compatibility testing, corrosion resistance, mechanical properties, ex-situ techniques, to name a few.

Development of models that include cost and technical performance (cost and performance): Similar to the state of tool development described above, models for RFCs and particularly metal/flow cells are underdeveloped compared to those for Li-ion batteries and PEM fuel cells. More important, these models are needed to guide many of the other development activities identified above. Models are needed for electrodes, cells, stacks, and systems. Again compared to other energy storage and conversion devices, scaling laws and parametric models are not available. Given the breadth of technologies being considered, a build and bust approach is impractical.

Identification of transport and kinetic properties (performance) Many of the physico-chemical properties needed for models and therefore design of electrodes are not available. Engineering research to develop methods of measuring transport properties and kinetic rate constants for the diverse systems being explored is needed. Further, these techniques and data should be organized and made available to the general community.

The key R&D priorities identified in these system included:

1. How to create uniform deposition and stripping of metal – avoiding dendrites, channel blocking
2. Create a stable colloidal dispersion of metal that can be charged and discharged
3. Development of highly loaded electrodes with equal performance
4. Development of models including cost and technical performance
5. Identification of transport and kinetic properties

Number 2 above is critical to enable the separation of energy and power that is a hallmark of RFCs.

Critical Topics	
Formation of the metal phase	<ul style="list-style-type: none"> • Plating • Dendrites on charge • Metal conditioning cycle
Side Reactions	<ul style="list-style-type: none"> • O₂, H₂ evolution • Passivation
Durability	<ul style="list-style-type: none"> • Carbon corrosion • Erosion • Seals
Separator/Membranes	<ul style="list-style-type: none"> • Solid state • Selectivity, permeability
Important Topics	
Diagnostics	
Electrode and cell design	<ul style="list-style-type: none"> • Metal loading • Gas holdup • Air electrode development • Activation
Independent cost analysis	<ul style="list-style-type: none"> •
System	<ul style="list-style-type: none"> • Scale-up • Controls • Shunt currents • BOP: slurry transport • Multiphase flow and rheological studies of dispersions
Modeling	<ul style="list-style-type: none"> • Transport properties and phenomena • Multiphase flow

B. Novel and nonaqueous systems – facilitator Reid Heffner, scribe Charles Monroe

The findings from this group focused on the applications for non-aqueous electrolytes including compelling reasons like the higher possible voltages (energy density) although the electrolytes will cost more. Energy and power density go up with high voltage and high concentration, so achieving reversible, energetic reactions involving high-solubility active species is clearly desirable. High power density reduces capital cost; high energy density doesn't, necessarily. But a high-energy system can outperform existing systems in terms of capital cost by lowering the necessary active-species loading.

One of the problems is that in these systems the "whole chemistry" is a variable. Thus, developing a roadmap to approach these barriers is difficult, particularly in an applied R&D effort. However, there are some fundamental questions such as: what are the rules of non-aqueous electrochemistry? And how does one produce viable non-aqueous membranes? Because of the vast parameter space, there is still a great need to identify and understand fundamentals of new systems (couples). Ways to do this include existing screening tools (e.g., voltammetry), and there is a need for quick screens for the possible couples. Ideally, electrode & electrolyte thermodynamic and stability databases and electrolyte transport measurements (conductivity, diffusion rates) are required. Good screening tools, both experimental and computational, are required. One key focus area was on the use of ionic liquids as the solvent\electrolyte as it is believed that their inherent safety and possibility for low cost outweigh those of other solvents. This area approaches the TRL 1 level due to the many unknown properties and compatibilities.

Key R&D needs included:

1. Electrolyte (e.g., ionic liquid) and electrode property databases
2. Rapid screening methodology
3. Electrode materials
4. Modeling and diagnostics
5. Sealing and dealing with organic solvents

Critical Topics	
Solvents (ionic liquids)	<ul style="list-style-type: none">• Low viscosity and rheologic properties for low pressure drops• Impurities (e.g., O₂)• Slurry flow• Plug flow• Sealing• Shunt currents
Electrode materials	<ul style="list-style-type: none">• Stability over large voltage range• Replacement of carbon instead of new ones?• Property/compatibility database• Mechanical properties

Modeling	<ul style="list-style-type: none"> • New modeling tools • Multiphase flow • Ion transport • Computational chemistry
Important Topics	
Separator/Membranes	
Diagnostics	<ul style="list-style-type: none"> • Electrochemical properties of bulk liquids • Transport property measurements
System	<ul style="list-style-type: none"> • Economic modeling compared to aqueous systems • Safety issues

C. Traditional single and multiphase systems – facilitator Michael Perry

The findings from this group focused on the needs for the community in terms of understanding the applications and developing roadmaps and metrics for the applied R&D.

The highest priority identified was the need to establish key requirements for at least some near-term markets. As noted, this is challenging because there are relatively large number of EES applications and markets. There are no actual round-trip energy efficiency targets for any of the applications. High round-trip energy efficiency is critical for applications that require frequent cycles and therefore store and release relatively large amounts of energy. However, there are other EES applications that do not require frequent operation since the value proposition is based on reducing the required capacity of the conventional grid (i.e., decreasing or delaying the need for either generation capacity and/or T&D capacity). In these “capacity applications” it is obvious that round-trip energy efficiency is less important than in “energy applications” and an EES device with relatively low energy efficiency combined with sufficiently lower capital cost (to offset the higher operating costs), may actually be more suitable. Capturing these trade-offs in key requirements is complex, so it is recommended that the best approach might be to have a simple EES Applications model (e.g., Excel based) that enables an EES end-user or EES developer to determine key technical requirements based on some basic input assumptions. The key input parameters would be technical requirements for the targeted EES application (e.g., discharge-time duration, response time, maximum discharge power, maximum charge power) and key outputs would be EES-device requirements (e.g., \$/kWh installed capital cost, round-trip energy efficiency), with the option of setting some of these outputs (e.g., \$/kWh) to determine others (e.g., round-trip energy efficiency). If this EES-application model was generic enough, it might also eliminate the need to generate requirements for each possible EES application, since users could use it as a starting point for the EES applications they wish to focus on.

Another high priority identified was establishing a baseline for current EES technologies, especially RFC systems. Currently, there are considerable discrepancies in this area and a third-party assessment of start-of-the-art (SOA) EES options would be valuable. *This would help to*

identify the major technical gaps that need to be addressed by comparing key requirements for targeted EES applications with the current options. These gaps, or needs, might be different for each type of RFC system, which is valuable to identify. For example, DOE's highest priority research needs would typically be those that address the gaps of multiple RFC technologies.

A key enabler to establishing the SOA of different RFC systems is to obtain performance metrics from units that have been deployed (i.e., field demonstrations). Key performance parameters measured in real-world applications (e.g., round-trip energy efficiency) are highly desirable and also probably easier to obtain from both the end-users and/or the developers than on laboratory systems. More importantly, *this data is especially valuable to determine major failure mechanisms, i.e., the reason(s) a unit removed from operation, which will help to identify key durability issues at an early stage.* Since establishing durability is always a lengthy process, especially for EES products that will typically require long lifetimes (e.g., > 10 yrs), it is absolutely critical to identify and address any durability issues as soon as possible. Fortunately, since RFC technologies were originally developed more than 30 years ago, there have already been some field demonstrations, but these results do not appear to be readily available.

All of the above are valuable goals, but they would also form the foundation for developing a Technology Roadmap with key targets for flow-cell technologies. These technical targets should be “SMART” metrics,¹ not just vague goals or project accomplishments that do not actually reflect technical progress. Specifically, key metrics for each major RFC chemistry should include:

- Theoretical energy density, expressed as kWh/L and kWh/kg
- Estimated cost, expressed as \$/kWh_{theoretical} (not \$/kg or \$/L as reported by Sandia)
- Maximum cell power density (W/cm²)
- Power density during discharge at a given efficiency (e.g., at approximately 70% cell voltage efficiency, which could be simply defined as OCV * 0.7)

These baseline metrics could be used as a starting point to define key RFC targets. Analogous to DOE's Hydrogen & Fuel Cell Program, these targets should become progressively more aggressive over time, and include interim targets to measure and track the program's technical progress. Additionally, the program should establish, clearly differentiate between, stack, system, and component levels targets. Currently, there are no targets established for RFC components (e.g., membranes), which makes it difficult for component suppliers to understand what is needed to advance RFC systems.

Somewhat analogous to fuel-cell systems, each major type of RFC system is defined by the chemistry of the electrolyte solutions. However, since there are two electrolyte solutions per system (i.e., a negative- and a positive-electrode solution, sometimes referred to as an anolyte and a catholyte), there is a wider variety of systems than encountered in fuel-cell systems. Therefore, DOE should probably not try to down select particular chemistries to begin with, but instead attempt to capture key properties for each of the possible reactant solutions. In addition to the key metrics listed in the section above, a more detailed electrolyte database would ideally

¹ SMART = Specific, Measurable, Actionable, Relevant, and Timely

tabulate some additional important physical properties for each type of reactant solution, such as viscosity, thermodynamic stability, solubility limits as a function of temperature, impurities, etc.

Key R&D needs include:

1. Develop targets for the various components (electrodes, electrolytes)
2. Develop a lessons learned database from the various installations and deployments
3. Understand the electrode design
4. Develop a physical property database for the various components with the various couples and solvents
5. Modeling of the cell and system
6. Understand the chemistry of the various constituents (e.g., carbon)

Critical Topics	
Redox couples (electrolytes)	<ul style="list-style-type: none"> • Modified by ligands to tune potentials • Supporting electrolytes and buffer systems • Electrolyte database of kinetic and thermodynamic properties • Material compatibility • Stability • Solubility limits • Qualified domestic suppliers that utilize low cost commodity materials
Durability	<ul style="list-style-type: none"> • Real-world data from demonstrations • Electrode corrosion and stability • Non carbon electrodes • Accelerated degradation and operating protocols • Cell starvation
Modeling	<ul style="list-style-type: none"> • Open system cost analysis of general systems • Complex modeling of the physical phenomena • Determine design targets at the cell and stack levels
Important Topics	
Codes and standards	
System	<ul style="list-style-type: none"> • No-shunt-current designs • Low pressure drops • Multiphase operation • Alternative structures including plug flow and micro channels

Breakout II

Individuals wrote down ideas on 3x5 cards, these were then each discussed and sorted to identify commonalities. From there, they identified those that were particularly important for RFCs and prioritized the research needs. Further, each idea was categorized based on diagnostics, materials, manufacturing, and modeling needs.

A. Catalysts/electrode designs – facilitator Fernando Garzon

The electrodes are critical components in RFCs. The electrodes need to be optimized for high electronic and ionic conductivity, high interfacial electrochemical charge transfer rates, efficient mass transfer of the reactant species, and low fluid drag to minimize pumping losses. Some of the RFC chemistries (such as oxygen cathode) require an electrocatalytic surface to promote rapid charge transfer. Other ones do not require a precious-metal surface; in fact, the supporting carbon fuel-cell diffusion media of paper, felts, or woven fibers makes ideal carbon porous electrodes. Of course, there is an issue of possible carbon corrosion at higher potentials, and so there is a need to develop understanding or mitigation methods for this corrosion, especially since the electrode materials must be corrosion resistant for tens of thousands of hours of operation and possess excellent dimensional stability. In addition, there are some changes as most RFC electrodes are flow-through and not flow-by, thus meaning that convection and mass transfer can be quite different. Bipolar, easily formable electrode materials lower the cost of the RFCs; however their use poses greater challenges in materials selection. The bipolar material needs to operate efficiently in both electrochemically oxidizing and reducing conditions and exhibit long-term durability. The development of cost effective RFCs for energy storage thus requires careful *co-optimization* of the desired anode and cathode properties.

New Electrode Materials Development: Many RFC technologies use graphitic carbons as relatively low cost electrode materials. Unfortunately carbon corrosion may be significant at potentials above 1 volt in aqueous acidic electrolytes and carbon corrosion may therefore be the limiting factor in RFC lifetime. The development of new carbonaceous and non-carbonaceous electrode materials research will foster the advancement of lower cost, higher performance, longer lifetime and higher efficiency batteries. Candidate material families may include:

- Novel forms of carbons i.e. fullerenes, nanotubes, nanowires and glassy carbons
- Advanced corrosion resistant metals and alloys
- Electronic conducting ceramics such as reduced oxides, nitrides and carbides
- Electronic conducting polymers
- Thin/thick film coatings of electrode materials on low cost supports
- New ultra high surface area materials

Optimization of Electrode Reactions: RFC technology is most valuable when the systems possess high volumetric energy storage densities and high volumetric charge and discharge power densities. An efficient way to achieve high volumetric power densities is to use electrodes which maintain high current densities at low overpotentials. The optimization of electrode performance requires a detailed understanding of the interfacial charge transfer processes and

kinetic rate limiting steps for the candidate chemistries. Novel electrode morphologic geometries may also enhance mass and charge transfer.

In battery electrochemistries that involve simple one electron outer sphere heterogeneous charge-transfer reactions, facile access to large charge transfer surface areas is a highly desirable characteristic. In these systems, it is often times difficult to probe the effective electrochemical surface area, particularly *in situ*, as the species are non-absorbing. New methods to characterize the electrochemical surface area and how this changes with battery operating conditions would be particularly useful.

Many battery chemistries require the adsorption and activation of the reactant species onto the electrode surface before appreciable electron transfer may occur. An extensive understanding of the heterogeneous and homogeneous phase electrocatalysis is vital to performance optimization. In some systems such as Fe/Cr, secondary mobile catalysts significantly increase the charge transfer rates. Measurements of surface species, calculations and measurements of the charge transfer rate as a function of applied potential will facilitate electrode modeling and optimization.

Electrode Degradation: Performance loss in RFCs is often times strongly coupled to electrode degradation through corrosion and morphological changes. To decrease electrode degradation it is imperative to study degradation rates and mechanisms for different carbons (graphitic amorphous, glassy, fullerenes) for each RFC chemistry; and degradation rates and mechanisms for new candidate materials, alloys, ceramics, polymers, etc.

Electrode Characterization: RFC performance has been for the most part, empirically optimized. Recent advances in electrochemical, surface and morphological characterization techniques can provide vital information on the performance and degradation mechanisms of RFCs. D.C. and A.C. electrochemical techniques can measure kinetic rates, corrosion reactions, surface areas, and probe transport processes. Advanced neutron, X-ray and NMR imaging techniques can visualize chemical and flow behavior in working cells. *In situ* spectroscopies can reveal mechanistic details of the electrochemical processes. Electron and X-ray microscopies can provide nano to mesoscopic morphological and chemical imaging of the electrode materials.

Modeling: The theoretical modeling of electrochemical processes has advanced greatly due to improved models, increased computational power and more detailed experimental validation. Theoretical modeling from the atomistic to the macroscopic level will help to improve system performance without the need for extensive prototyping and improve the understanding of the mechanisms of failure and efficiency losses.

The key R&D priorities identified in these system included:

1. Material development, especially for non-carbon electrodes
2. Optimization of the electrode reactions
3. Electrode degradation
4. In-situ and ex-situ characterization, especially determination of a reproducible and accurate measurement of the electrochemically active surface area
5. Modeling

Critical Topics	
Novel electrode materials	<ul style="list-style-type: none"> • Corrosion resistant • Non carbon • Electrocatalysts including immobilization • Nanostructure and novel geometries • Optimized for both charging and discharging
Diagnostics	<ul style="list-style-type: none"> • Measurement of surface area • Kinetics and reaction mechanisms • Establish or borrow from fuel cell ex- and in-situ techniques • Chemistry related techniques
Degradation	<ul style="list-style-type: none"> • Establish life targets
Important Topics	
Microfluidics and liquid transport	<ul style="list-style-type: none"> • Fluid friction at the nanoscale • Interface boundary layer friction in the presence of large compositional gradients • Cell designs with flow pressure drop but high mass transfer
Modeling	<ul style="list-style-type: none"> • Couple with performance • Validate • Determine controlling reaction steps
Establish technical targets	

B. Membranes – facilitator Thomas Zawodzinski

Membranes are one of the key components of both fuel-cell and RFC systems, where for fuel cells one worries about efficient proton conduction and limited crossover, whereas the interaction in RFC electrolytes is much more complicated due to the presence of many ions. This also results in a situation where crossover becomes easier and thus is more critical to manage, similar to direct-methanol fuel cells.

Discussion focused a lot on how to organize the effort in terms of something like centers of excellence. There was also a sentiment that other DOE programs should be leveraged including possibly solar and the advanced manufacturing. Key R&D priorities included:

1. Understanding the multicomponent transport inside the membranes including diagnostics and modeling

2. Material development and different compositions and chemistries and their requirements for the different applications
3. Membrane degradation
4. Standards

Critical Topics	
Chemistry	<ul style="list-style-type: none"> • Fluorinated vs. hydrocarbon • Composite and reinforced or bilayer membranes • Solid-state ion conductors
Durability	<ul style="list-style-type: none"> • Chemical properties • Mechanical properties • Impurities and fouling • Degradation mechanisms
Transport	<ul style="list-style-type: none"> • Quantify transport properties (i.e., conductivity, selectivity, cross-over) • Develop a set of careful, well-defined diagnostic measurement methods • What are we transporting? • Transport/parasitic loss understanding • Modeling • Non-aqueous especially
Important Topics	
Standards	<ul style="list-style-type: none"> • Technical targets for the various applications • Baseline materials including both porous and nonporous membranes
Examine manufacturing issues	

C. Stack, system, and related components – facilitator Ned Stetson, scribe Kathy Padro

The key issue for cells and stacks are that the subscale experiments must be representative of full-size-cell behavior. The objective is to improve performance with lower cost materials, which also means that the materials must be very durable under the conditions within the RFB chemistry of interest. In stack designs, issues of shunt currents are very prevalent and this must be accounted for. In addition, the problem of mass transfer and pumping losses of liquid electrolytes can become very energy inefficient and thus alternative stack and cell designs may be warranted. Thus, there is a tradeoff between balance of plant (e.g., pumps and pumping losses) and number of cells and shunt currents that needs to be understood. Similarly, there is also a tradeoff between cell size and count and power electronics as the RFC designer would like higher currents while the power-electronics engineer prefers higher voltages.

The key R&D priorities included:

1. Bipolar plates including shunt breaks to prevent shunt currents, new designs, non-carbon materials to alleviate cost but not necessarily weight, stability
2. Power electronics where there is a tradeoff between cell size, count, and the power-electronics equipment
3. Novel stack designs, especially ones that can mitigate shunt currents and/or pumping losses
4. Seals and gasket materials including compatibility and durability, especially since the liquid electrolyte solvents are often hard to seal (e.g., KOH)
5. Storage technologies
6. System modeling

Critical Topics	
Bipolar plates	<ul style="list-style-type: none"> • Noncarbon and robust • Composite design • Understand duty cycle • Compatibility and corrosive stability
Storage	<ul style="list-style-type: none"> • Stability and electrolyte dependent • On-site fabrication • Compression • Modeling
Other materials and compatibility	<ul style="list-style-type: none"> • Seals • Gaskets • Sensors
Important Topics	
Power electronics	<ul style="list-style-type: none"> • Capital vs. operating cost trade • Power quality vs. performance tradeoff
Determine quality assurance metrics	<ul style="list-style-type: none"> • Electrolytes • Pumps, pipes and valves • Materials of construction

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Appendix A: Workshop Agenda

Day/Time	Speaker	Subject
Wednesday, March 07, 2012		
8:45-9:00	Adam Weber, LBNL	Welcome and workshop overview
9:00-9:30	Sunita Satyapal and Dimitrios Papageorgopoulos, EERE, FCTP	Background, approach, and reversible fuel cells
9:30-9:55	Michael Perry, UTRC	Renaissance in flow cells: opportunities
9:55-10:20	Joe Eto, LBNL	Energy storage requirements for the smart grid
10:20-10:35	Break	
10:35-11:00	Robert Savinell, CWRU	Revisiting flow-battery R&D
11:00-11:25	Stephen Clarke, Applied Intellectual Capital	Lessons learned and yet to be learned from 20 years in RFB R&D
11:25-11:45	Imre Gyuk, DOE OEDER	Research and deployment of stationary storage at DOE
11:45-12:05	Mark Johnson, DOE ARPA-E	Flow cell research in GRIDS
12:05-12:30	Tom Zawodzinski, ORNL and UTK	Transitioning fuel-cell technology to RFBs
12:30-1:30	Working Lunch and breakout group instructions	
1:30-3:30	Breakout Groups I	A. Metal/flow systems B. Novel and nonaqueous systems C. Traditional single and multiphase systems
3:30-3:45	Break	
3:45-5:00	Reports from Breakout Groups I	
5:00-5:15	Summary of Day 1	
5:30-7:00	No-host reception	
Thursday, March 08, 2012		
8:30-8:45	Breakout group instructions	
8:45 - 10:45	Breakout groups II	A. Catalyst/electrode design B. Membranes C. Stack, systems, and related components
10:45-12:00	Reports from Breakout groups II	
12:00-12:15	Workshop Summary	

Appendix B – GRIDS Program Targets

<https://arpa-e-foa.energy.gov/Default.aspx?Archive=1#FoaId85e239bb-8908-4d2c-ab10-dd02d85e7d78>

PRIMARY TECHNICAL REQUIREMENTS:

Requirement ID Number	Requirement Category	Value (Units)
1.1	System Capital Cost per Unit of Rated Energy Capacity (for measured capacity at Rated Power)	<\$100/kWh
1.2	Minimum Operating Time at Rated Power (time at Rated Power for charge and discharge)	60 minutes
1.3	Maximum Response Time (time for system to go from 0% to 100% of rated power in discharge and in charge mode)	10 minutes
1.4	Rated Power Capacity for Charge and Discharge in Advanced System Prototypes	≥20kW

SECONDARY TECHNICAL TARGETS:

Target ID Number	Target Category	Description
2.1	Cycle Life (cycled at rated power between charge and discharge)	5,000 cycle minimum, defined as number of cycles at which >20% reduction in total energy/power capability occurs relative to initial rated values
2.2	Round-Trip Efficiency	80% at rated power for of charge and discharge
2.3	Maximum Dwell Time	Maximum 10 minute response time for reversal between charge and discharge cycles
2.4	Scalability of Storage Technology for Grid-scale Application	Potential for subsequent scaling for grid-scale deployment (1-10MW). Scalability will be assessed at the power/energy ratio of the advanced systems prototype proposed.
2.5	Internal Losses	Less than 5% loss of energy in 24 hours from fully charged state.
2.6	Safety	Consistent with transmission and distribution grid deployment at unattended locations
2.7	Calendar Life	10 years minimum