

CHAPTER 4

SODIUM-BASED BATTERY TECHNOLOGIES

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Abstract

The growing demand for low-cost electrical energy storage is raising significant interest in battery technologies that use inexpensive sodium in large format storage systems. Potentially viable candidate technologies today include relatively mature molten sodium batteries and emerging sodium ion batteries. While still relatively expensive, molten sodium battery chemistries, such as sodium-sulfur (NaS) and sodium-nickel chloride (Na-NiCl₂), are technologically mature enough for global deployment on the scale of hundreds of megawatt-hours. (MWhs). Significant applications of these technologies include renewable integration, backup power, and additional grid services while meeting demands for consumer, commercial, and industrial energy stakeholders. As research and development efforts continue in academia, national laboratories, and industry, widespread use of safe, cost-effective molten sodium batteries as well as implementation of new sodium ion-based batteries are expected to be important elements of the evolving energy storage community.

Key Terms

Grid-scale storage, sodium (Na), sodium-ion battery (NaIB), sodium metal, sodium-nickel chloride (Na-NiCl₂), sodium-sulfur (NaS)

1. Introduction

Sodium is the sixth most abundant element on Earth, it is widely distributed globally, and it is already processed on large scale as an industrial material, making it an attractive constituent for cost-effective, large-scale energy storage. Commercially-relevant sodium batteries today can be roughly grouped into two primary classes: molten sodium batteries and sodium-ion batteries. Both approaches to sodium utilization are discussed here, though the commercialization and deployment of molten sodium batteries is presently more advanced than that of the sodium-ion systems.

1.1. Molten Sodium Batteries

Research and development of molten sodium batteries began with the sodium-sulfur (NaS) battery in the late 1960s, followed in the 1970s by the sodium-metal halide battery (most commonly sodium-nickel chloride), also known as the ZEBRA battery (Zeolite Battery Research Africa Project or more recently, Zero Emission Battery Research Activities). Schematically depicted in Figure 1, both systems take advantage of a molten sodium anode and a ceramic sodium-ion conducting solid state separator, most commonly β ''-alumina (BASE [Beta Alumina Solid Electrolyte]), but the molten cathode chemistry in each case differs significantly, as described below.

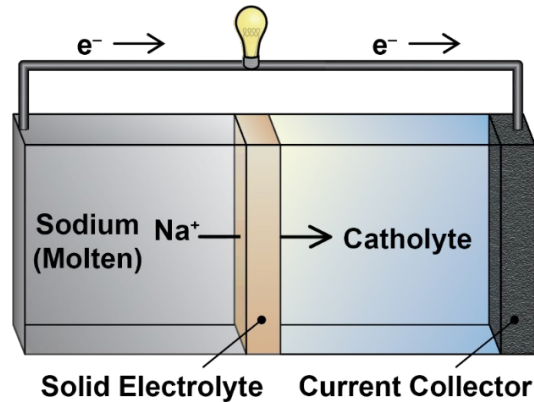


Figure 1. Schematic illustration of a molten sodium battery

1.2. Sodium-Sulfur (NaS) Batteries

During electrochemical cycling of the batteries, NaS batteries oxidize (discharge) and reduce (charge) sodium, relying on the reversible reduction (discharge) and oxidation (charge) of molten sulfur. During a typical discharge reaction, oxidized Na^+ crosses from the anode through the ion-conducting ceramic separator and reacts with the molten sulfur (or polysulfides) reduced at the cathode. This reaction produces molten polysulfides (e.g., Na_2S_5) that can be electrochemically cycled.



At values of $x < 3$, insoluble (solid) polysulfides form, degrading battery performance. As a result, these batteries are limited to approximately half their theoretical capacity. The low cost of sulfur as a reagent, however, means that it is reasonable to overbuild the cathodes to avoid capacity limitations.

1.3. Sodium-Nickel Chloride (Na-NiCl₂ or ZEBRA) Batteries

ZEBRA batteries, like the NaS battery, rely on the oxidation and reduction of sodium at the anode and use a BASE separator, but they rely on the oxidation and reduction of nickel metal at the cathode. Specifically, the cathode reaction is supported in a metal halide molten salt electrolyte, traditionally NaCl (sodium chloride) and AlCl_3 (aluminum trichloride), which combine to form NaAlCl_4 (sodium tetrachloroaluminate). As such, these batteries are also referred to as sodium-metal halide batteries or molten salt batteries, or even just salt batteries. The overall electrochemical reaction of this system is:



In this case, the cathode chemistry does not remain entirely liquid; the electrochemical cycling results in the formation of solid (nickel chloride-coated) nickel particles and even solid sodium chloride particles. These batteries are also operated near 300°C to maintain high conductivity of the BASE separator and to ensure the molten state of the molten salt catholyte. At these high temperatures (near $270\text{--}300^\circ\text{C}$), there are concerns about the potentially corrosive nature of the molten salts, though they are less corrosive than the sodium polysulfides found in NaS systems. At high temperatures, there is also an issue associated with irreversible growth of solid phase particles (coarsening) during electrochemical cycling, which can lead to loss of capacity over time [1].

1.4. Sodium-ion Batteries (NaIBs)

Although molten sodium batteries continue to grow as a relatively mature technology, sodium-ion batteries (NaIBs) are making advances toward large-scale commercialization. Many developers envision NaIBs as safer alternatives to lithium-ion (Li-ion) batteries, including applications for portable electronics and vehicle electrification. In this chapter, however, the focus will remain on system development for grid-scale applications. NaIBs operate differently from the molten sodium batteries described above, with their electrochemistry more closely resembling that of a Li-ion battery. These batteries typically operate at room temperature and employ a transition metal cathode, a non-selective, electrically insulating porous polymer separator, a carbon or a titanate anode, and an organic or aqueous liquid electrolyte. Battery function involves alternately intercalating sodium ions into the cathode during discharge and the anode during charge. Unlike the molten sodium batteries mentioned previously, the separators do not need to be selective solid-state materials; these batteries can use the same types of porous polymer materials used in more mature Li-ion technologies. The separator primarily serves to prevent physical contact and electrical shorting between anode and cathode.

Despite the similarities to Li-ion batteries, there are a few important distinctions between these systems and their Li-ion counterparts. First, they are generally considered safer than Li-ion systems, primarily due to the use of less volatile electrolytes and different oxides for the cathode. In addition, some NaIBs can be fully discharged, allowing the system to be safely transported, for example, by air. The NaIB cathodes are also different from those of Li-ion systems, and they often do not rely on the use of cobalt (Co) or nickel (Ni), two potentially expensive and geopolitically limiting components of many Li-ion systems. The carbon anodes in these systems are also different from the graphitic carbons favored in commercial Li-ion batteries as these do not accommodate significant Na-ion insertion. Instead, amorphous “hard carbon” is frequently used in NaIBs. Finally, due to the larger size of the sodium ion compared to the lithium ion, and the slightly smaller reduction potential of sodium, it is generally expected for NaIBs to be less energy dense than their Li-ion counterparts, though this metric can be accommodated for by different degrees of ion intercalation and battery engineering. Importantly, it is reasonable to expect that despite these subtle differences in battery composition, NaIBs could be potentially manufactured on large Li-ion production lines.

2. Current State -of-the-Art

Currently, only NaS and Na-NiCl₂ systems are commercially mature grid-scale technologies [2-3]. Both of these technologies are built as modular units that can be scaled to store energy from a few kilowatt-hours (kWhs) to tens of megawatt-hours (MWhs) of energy. These systems do not employ the simplified “planar” battery design shown in Figure 1. For both commercial systems widely used today, the manufacturers use closed-end tubular (often clover-leaf cross-sections) separator designs. In the case of the “NAS” system developed by NGK Insulators, Ltd. (Figure 2), the sodium anode is placed inside the ceramic separator tube and the sulfur cathode surrounds the exterior of the tube [3]. The system, complete with current collectors is sealed and packaged in an elongated cell (Figure 2, left). Many of these cells are then packed together into a module, and modules are packed into larger, scalable containers that are installed and controlled through an external power conversion system. These designs simplify battery assembly and operation while providing scalable high-packaged energy density.

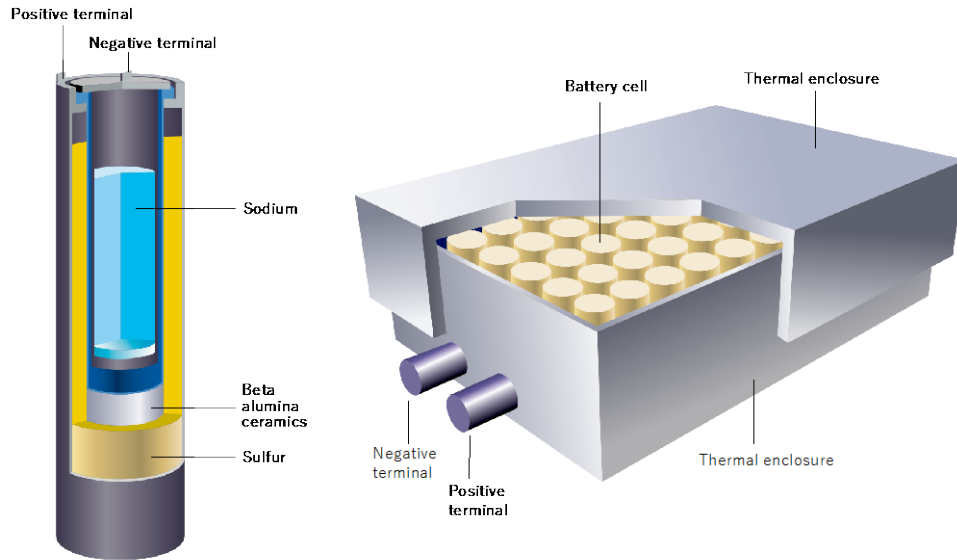


Figure 2. Illustration of a tubular battery design used for sodium sulfur batteries. The tubular cell assemblies are packaged and connected in a thermal enclosure to create functional modules. Images provided courtesy of NGK Insulators, Ltd.

Key metrics for these batteries are summarized in Table 1. As these values indicate, NaS systems are valued for high energy density, scalability (tens to hundreds of megawatts), their ability to discharge (rated power) over the course of 6-7 hours with rapid response times (less than 1 second), and reasonable round trip energy efficiencies. As robust, self-contained systems, they are considered “maintenance-free” over the course of 10-15 years, with expectations of 4,000-4,500 total cycles (80% depth of discharge (DOD)). (NGK expects 300 cycles per year for 15 years [3].)

Table 1. Comparison of Metrics Typical of Commercial Molten Sodium Batteries. Values taken from Refs 1-3.

	Practical Energy Density (Wh/L)	Expected Cycle Life (cycles at 80% DOD)	Expected Operational Lifetime (years)	Operating Temperature (°C)	Discharge Duration (at rated power)	Round-Trip Efficiency
NaS	300-400	4,000-4,500	15	300-350	6-7 hours	80%
Na-NiCl₂	150-190	3,500-4,500	20	270-300	2-4 hours	80-85%

2.1. Sodium-Sulfur Batteries

Until now, the widespread deployment of NaS batteries has been limited by a few select factors. First, these batteries operate near or above 300°C, which ensures not only the molten character of the sodium and the sulfur/polysulfide catholyte, but also the low ionic resistance of the BASE separator. Counterintuitively perhaps, because the systems are engineered to control and maintain this elevated temperature efficiently, they are functional across a wider range of environmental temperatures than most other technologies (-20°C to +40°C). Less desirably, however, at the high operating temperatures, the sulfur/polysulfide catholyte is strongly corrosive, and in the event of a

separator failure, reaction of the molten sodium with the sulfur catholyte can result in a toxic, highly exothermic fire capable of inducing thermal runaway in a large-scale system. Great efforts have been put toward engineering housing materials, their coatings, and sealing materials that are resistant to such corrosion along with additional cell designs to further limit exothermic runaway conditions in the event of a separator failure or short circuit. In fact, since a “fire incident” at the NAS Tsukuba Plant (Josu City, Ibaraki Prefecture, Japan) in September of 2011 [4], there have been no recognized large-scale fires from this technology. Unfortunately, the engineering solutions required to address the high temperature materials requirements and the improved safety of the system contribute to a relatively high cost of these batteries. Current costs are estimated between \$600-700 /kWh with projections that costs could drop to less than \$500 /kWh by 2025 [5]. Although these “up-front” capital expenditure (CAPEX) costs are relatively higher than current Li-ion or lead-acid (Pb-acid) alternatives, the long duration discharge, minimal maintenance, and extended cycle life of these systems should make them more attractive for long-term applications where the costs can be amortized over the lifetime of the system.

2.2. Sodium-Nickel Chloride Batteries

As summarized in Table 1, Na-NiCl₂ batteries share many of the positive attributes of the NaS systems, with favorable energy densities, long cycle lives, long calendar lives with little or no maintenance, and good round trip efficiencies. Again, although these batteries operate near 270°C, they can be used in environments with significantly variable ambient temperatures (-40°C to +60°C) [2]. Importantly, though, Na-NiCl₂ batteries are considered to be “inherently” safer than their NaS counterparts; reactions between the molten salt and the molten sodium anode do not yield runaway exothermic behavior, produce toxic byproducts, or generate hazardous volatile species. Battery manufacturer FZSoNick recently announced that their Na-NiCl₂ battery systems (Figure 3) were UL 9540A certified (Test Method for Evaluating Thermal Runaway Fire Propagation in Battery Energy Storage Systems), attesting to their relative inherent fire safety. FZSoNick also advertises that these batteries are considered to be non-hazardous and fully recyclable. It should be noted that molten sodium batteries, as a class, often elicit concerns related to the strong reactivity of metallic sodium with air and water. These concerns can be significantly addressed, however, through proper packaging and sealing of the cells to prevent environmental exposure. Moreover, for issues such as battery assembly or shipping, large volumes of metallic sodium need not necessarily be handled if the batteries are assembled and/or shipped in the discharged state.



Figure 3. An FZSoNick 48TL200 sodium–nickel battery with welding-sealed cells and heat insulation. 2012 Image by: Rudolf Simon–Own work, CC BY-SA 3.0. (<https://commons.wikimedia.org/w/index.php?curid=18740786>)

Like the NaS systems, the relatively high cost of Na-NiCl₂ batteries has limited their widespread distribution. The high temperature operation still introduces more expensive materials and engineering requirements, and the relatively high cost of nickel keeps costs higher than other technologies. They are currently manufactured and distributed by the FZSoNick, which maintains manufacturing capacity for over one million cells per year [2]. Estimated costs for these batteries are around \$700/kWh, with projections that these costs may drop to less than \$500/kWh by 2025 [5]. General Electric Company originally developed a ZEBRA battery product, known as Durathon, but this technology was never significantly distributed commercially. The Chinese energy storage company Chilwee recently acquired the Durathon technology and has announced plans to begin manufacturing these batteries as part of a more comprehensive battery manufacturing effort. The Fraunhofer Institute for Ceramic Technologies and Systems in Germany has also developed their own Na-NiCl₂ battery platform (Cerenergy) for grid-based energy storage [6]. They advertise an effort to adapt the cell design and improve materials chemistry in these systems to reduce cost, but at present, these systems are not widely deployed.

2.3. Sodium-Ion Batteries

Although not yet considered commercially mature technologies for grid-scale applications, there are several variations of NaIBs that are currently in development or production. The technologies described here are making significant progress toward implementation, but these do not represent a complete list of all NaIB technologies in development in academia, national labs, and emerging industry. Here, we briefly discuss NaIBs using Prussian blue analogs (PBAs) as electrode materials, batteries using electrode materials directly analogous to those used in Li-ion batteries, and batteries using aqueous electrolytes.

PBAs are materials composed of ferric ferrocyanide salts that are predominantly used as cathodes, though they can be used as anode materials as well. PBAs consist of a cubic crystal structure with large channels to accommodate the rapid movement of Na^+ ions in and out of the material with minimal volume change. This attribute grants these materials the advantage of high specific capacity, high rate capability and high cycling stability. Furthermore, they present low manufacturing costs due to the aqueous coprecipitation method necessary for production. Despite the presence of the cyanide ion, PBAs are largely non-toxic as the cyanide is tightly bound to the iron. Their main disadvantage, a low crystal density, is acceptable for select grid storage applications where volumetric energy density can be relatively low [7]. California-based startup, Natron Energy, is currently developing NaIBs that use different grades of PBAs, with deployments of 8 kW units for data server backup power [8].

Li-ion battery analogs are a popular area of development, owing to the potential for rapid deployment and low CAPEX by the use of existing large Li-ion production lines. One company, China-based HiNa Battery Technology, has leveraged this to produce over 10,000 prototypes within 2 years of being founded, and is currently demonstrating a 100 kWh NaIBs system with a pyrolyzed anthracite anode and a Co- and Ni-free layered transition metal oxide cathode [9]. UK-based Faradion, one of the earliest NaIB startup companies, has succeeded in producing over 50 kWh of prototypes with their sodium-nickel layered oxide cathode since their founding in 2011 [10].

NaIBs using aqueous electrolytes are particularly attractive for grid storage applications, owing to the lower cost and higher safety attributed to the aqueous electrolyte, as well as the faster ionic transport within aqueous media. Such systems boast environmentally benign chemistry, often being referred to as “saltwater batteries.” Aquion Energy, a Pennsylvania-based company, succeeded in the worldwide deployment of their aqueous “Aspen” battery systems on the MW scale, before filing for Chapter 11 bankruptcy and restructuring in 2017. Their system used a carbon titanium phosphate composite anode, sodium perchlorate aqueous electrolyte, and manganese oxide cathode. A very similar system (Greenrock Saltwater Battery) appears to be marketed by BlueSky Energy out of Austria [11].

NaIBs are a relatively new technology in the grid-scale energy storage space and are primarily in the research and development and demonstration stages. A large number of companies are taking the initial steps towards bringing these batteries to market, and those mentioned earlier are not a comprehensive listing. Demonstrations for grid storage, such as backup power systems and energy arbitrage for microgrids, are ongoing and provide optimism that NaIB technologies will provide another piece of the energy storage puzzle in the near future.

3. Grid-Scale Battery Deployment

The attractive high performance, long cycle life, and minimal maintenance of molten sodium batteries led to significant deployment of these systems globally. In general, these batteries are suitable for load shifting, peak shaving, frequency regulation, renewables integration, voltage control, and backup power, though there may be additional applications viable that take advantage of their fast response times and long-duration storage.

3.1. Sodium-Sulfur Battery Deployments

At present, NGK appears to be the only significant manufacturer of the NaS technology. As of early 2020, NGK reports 560 MW/4,000 MWh of storage in more than 200 locations around the world, with some of these installations operating successfully for over a decade [3]. The applications using this technology vary significantly, addressing industrial, commercial, and residential storage needs. Specific areas of current utility include renewable stabilization, investment deferral and ancillary services, and powering microgrid and remote or island communities.

As examples of the NaS deployments, large installations support the integration of renewable wind and solar assets in Japan. A 34 MW/245 MWh system supports a 51 MW wind farm in Rokkasho, Aomori, Japan and a 50 MW/300 MWh system supports a large solar array in Fukuoka, Kyusyu, Japan (Figure 4). The Fukuoka solar installation is the largest NaS system in the world and it helps balance intermittent solar energy supply and variable energy demand. While the Fukuoka installation is relatively new (2016), the Rokkasho NaS system has been a flagship project operating to balance supply and demand from intermittent wind generation successfully since 2008.



Figure 4. A 50 MW storage system supporting a solar array in Fukuoka, Kyusyu, Japan. Image provided courtesy of NGK Insulators, Ltd. (<https://www.ngk-insulators.com/en/product/nas/solutions/ancillary/index.html>)

For islands and microgrid applications, NGK's NaS batteries are not only used to integrate and stabilize energy from local renewable resources (e.g, in Reunion Islands, France) but they have additionally demonstrated support for microgrid operation on Catalina Island, CA, United States. This 1 MW system provides alternative power in support of a diesel-powered energy grid when demand pushes diesel generation outside allowable operational ranges (80-100% capacity).

In Italy, NGK is working with the transmission system operator Terna S.p.A. as part of an effort to integrate renewable wind energy. NaS batteries, capable of deep cycling, have been able to facilitate long-distance electricity transmission by providing a storage buffer during periods of transmission congestion (Figure 5).



Figure 5. An NAS battery installation in the Campania region of Italy is used to facilitate renewable energy transmission. Images courtesy of NGK Insulators, Ltd. (<https://www.ngk-insulators.com/en/product/nas/>)

These are only a few examples of how NaS systems can enable grid-scale storage applications. As demand for these storage capabilities grows and research and development helps lower battery costs, increased deployment may be anticipated.

3.2. Sodium-Nickel Chloride Battery Deployments

FZSoNick continues to deploy their current, modular systems for a number of large-scale storage applications, with batteries suitable for frequency regulation, load shifting, peak shaving, backup power, and renewables integration [2]. They categorize their technology space into energy backup (48V-110V modules), mobile applications (vehicles) (300V-700V modules), and energy storage (48V and 620V modules). The vehicle applications are not a focus of this Handbook, but it is worth noting that these systems are sufficiently safe and have adequate energy density for select vehicle applications.



Figure 6. Images of 48V and 620V FZSoNick battery modules and their scalable integration into deployable containers. Reprinted with permission of FZSoNick.

FZSoNick has used Na-NiCl₂ systems to provide backup power for telecommunications, public transportation, and remote site applications. At present, FZSoNick has deployed roughly 100 MWh of energy storage with approximately 2 MAh of backup storage capacity, most heavily focused around telecommunications backup. They have deployed more than 14 MWh of additional storage for use in “Energy Storage Systems” that enable renewables integration, microgrid applications, and grid services (e.g., grid balancing, voltage regulation) [2].

The backup energy market is where these batteries have found their most significant application as a large-scale system, with applications in North America, South America, Europe, and to a

lesser degree, Asia. For example, in the United States, Na-NiCl₂ batteries provide stand-alone backup power for telecom data centers at 17 sites for two major cell phone providers. In Mexico, Zambia, and the Philippines, Na-NiCl₂ batteries provide backup power for telecom systems.

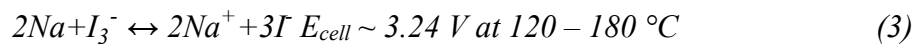
For larger-scale energy storage systems, Na-NiCl₂ batteries are used in the Maldives Islands (1.2 MWh), Ollagüe, Chile (560 kWh), Tilos, Greece (2.88 MWh), and French Guiana (4.5 MWh) to support integration of renewables such as photovoltaics and wind power. In a program similar to that described earlier for NGK's NAS systems, FZSoNick is also engaged with Terna S.p.A., using a 4.5 MWh system to facilitate electricity transmission in Sicily and Sardinia.



Figure 7. An FZSoNick Na-NiCl₂ battery system supporting integration of renewable remote photovoltaic energy in South America. Reprinted with permission of FZSoNick.

4. Emerging Efforts

Although both NaS and Na-NiCl₂ batteries are currently deployed as grid-scale technologies, research and development efforts continue to push for advances that would reduce cost and improve the safe, reliable performance of molten sodium batteries. Pacific Northwest National Laboratory (PNNL) has a cooperative research program with South Korean partners Korea Institute of Energy Technology Evaluation and Planning (KETEP) and Research Institute of Industrial Science and Technology (RIST) developing ZEBRA batteries that operate below 200°C and reduce or eliminate the use of costly nickel in the cathode, replacing nickel with iron [1, 12]. Researchers at Sandia National Laboratories are actively developing alternative low to intermediate temperature batteries that use a molten sodium anode and a NaI-based metal halide molten salt cathode [13]. The electrochemistry of these batteries relies on the oxidation or reduction of iodide at the cathode and the overall battery chemistry is:



This approach results in an increased cell voltage, relative to NaS or Na-NiCl₂ chemistries, (enabling potentially increased energy density) and allows for operation at temperatures near the melting temperature of sodium (97.8°C) [14].

Both of these national laboratory-led efforts use chemistries designed for safety (not thermal runaway) and are clearly aimed at reducing the operating temperature of these batteries, which would reduce or eliminate the need for high temperature stable battery components, reduce costs associated with maintaining battery operational temperatures, and decrease degradation or side reactions deleterious to long-term battery performance. Ultimately, finding lower temperature solutions will enable more widespread adoption of this class of batteries, but neither of these low-to-intermediate approaches have yet reached commercial technical maturity.

There have been several reports proposing to develop molten sodium batteries that use aqueous or organic catholytes to access lower temperature applications. While these systems may provide some important academic insights into battery performance, the potentially violent reactivity of the molten sodium with these materials in the event of separator failure makes them less likely candidates for commercial development. Still, if this issue can be mitigated, these may also be viable technologies in the future.

5. Summary and Conclusions

Large-format, grid-scale sodium-based batteries can take a number of forms, using both molten sodium chemistries and varied sodium-ion chemistries. Although Na-ion chemistries are rapidly becoming increasingly commercially relevant, current mature technologies include molten sodium-sulfur (NaS) and molten sodium-nickel chloride (Na-NiCl₂). Both systems use domestically abundant sodium metal anodes, ceramic ion conducting separators, and electrochemically active molten (or molten suspension) cathodes. Each of these chemistries operate at elevated temperatures, near 300°C, which requires select battery material candidates and system designs that result in relatively high cost. These batteries are, however, expected to provide rapid response times, hours-long discharge durations, deep cycle discharge, and long cycle lives over decades of low-maintenance use. These attributes have enabled two established primary battery manufacturers, NGK Insulators and FZSoNick, to deploy hundreds of MWhs of NaS and Na-NiCl₂ battery storage, respectively, around the world. Suitable for load shifting, peak shaving, frequency regulation, renewables integration, voltage control, and backup power, these batteries have enabled a wide range of industrial, commercial, and residential energy storage projects. Continued growth in demand and emerging innovations in both molten sodium and sodium-ion battery technologies promise new opportunities for sodium batteries to advance global energy storage.



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