Low-Temperature Multielement Fusible Alloy-Based Molten Sodium Batteries for Grid-Scale Energy Storage

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ABSTRACT: The sustainable future of modern society relies on the development of advanced energy systems. Alkali metals, such as Li, Na, and K, are promising to construct high-energy-density batteries to complement the fast-growing implementation of renewable sources. The stripping/deposition of alkali metals is compromised by serious dendrite growth, which can be intrinsically eliminated by using molten alkali metal anodes. Up to now, most of the conventional molten alkali metal-based batteries need to be operated at high temperatures. To decrease the operating temperature, we extended the battery chemistry to multielement alloys, which provide more flexibility for wide selection and rational screening of cost-effective and fusible metallic electrodes. On the basis of an integrated experimental and theoretical study, the depressed melting point and enhanced interfacial compatibility are elucidated. The proof-of-concept molten sodium battery enabled by the Bi−Pb−Sn fusible alloy not only circumvents the use of costly Ga and In elements but also delivers attractive performance at 100 °C, holding great promise for grid-scale energy storage.

As one of the most essential ingredients in modern economies, energy plays a critical role in the progress and evolution of human society. Considering the intermittent nature of renewable sources, advanced energy-storage systems are greatly needed to achieve efficient harvesting and delivery of the generated electricity.1,2 Up to now, pumped hydro, where energy is stored in the form of gravitational potential energy of water, is the most widely used technology for grid energy storage.3 Despite the relatively high efficiency, pumped storage hydroelectricity is geographically restrained, and there are also some environmental and ecological issues caused by the frequent change of water levels. Therefore, rechargeable batteries, in which electrical energy can be converted to chemical energy, are gaining more and more attention as a promising energy-storage technology to better regulate the electricity in peak load shifting.4–5 Among different types of electrochemical energy-storage devices, liquid metal batteries offering both high energy densities and high power densities stand out for stationary storage.6,7 Different from solid-state counterparts, liquid metal electrodes are immune to structural cracks, and the intrinsic dendrite growth of alkali metals can also be circumvented thanks to the self-healing feature.8–12 Meanwhile, superior power capabilities are expected given the fast charge kinetics in liquids. The liquid metal battery prototype can be traced back to the 1960s, and the battery chemistry has been extensively studied since then with combinations of various electrodes and electrolytes.13–20 Although significant progress has been made, high operating temperatures are typically needed to melt the metallic electrodes, which impede the wider implementation.21

The melting point of a substance can be roughly quantified by the intermolecular forces between atoms.22 Metals, generally speaking, tend to have high melting points considering the intense bonds originated from the strong electrostatic attraction forces between metal ions and free electrons. Accordingly, state-of-the-art liquid metal cells, relying on pure metal-based electrodes, need to be operated at elevated temperatures to maintain the molten state of electrodes and high conductivity of electrolytes.6,21 Different from pure metals, the melting points of metallic alloys can be reduced remarkably, especially in cases of eutectics, showing great feasibility for liquid metal battery application. On the one hand, the introduction of metallic heteroatoms will induce disorders in the alloy structure, which is beneficial to lowering the melting points.23 On the other hand, the localized charge density around metal atoms will be altered in the presence of adjacent heteroatoms, and such free-electron density difference contributes to depressed melting points in the meantime.24 For example, the operating temperature of liquid metal batteries has been decreased from 500 to 240 °C when replacing Pb with Pb−Sn or Pb−Bi alloy-based cathodes.21 Nevertheless, the widespread application of liquid metal batteries is still restrained by the sophisticated cell structure, rigorous thermal

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management, and potential safety issues, all of which are related to the relatively high operating temperature. Although Ga-based alloys can be adopted as electrodes for room-temperature liquid metal batteries, the high cost of Ga restricts its practical applications. A ternary alloy system was also reported to build liquid metal batteries, but the operating temperature is still as high as 500 °C. To further decrease the operating temperature of liquid metal batteries, we extended the battery chemistry of liquid metals to multielement fusible alloys. Compared with the unary metals, the multielement alloy system possesses more flexibility for wide selection and rational screening of cost-effective and fusible metallic electrodes to design liquid metal batteries. Herein, a series of Bi-based fusible alloys were systematically studied, namely, Field’s metal (Bi–Sn–In eutectic alloy, melting point of 62 °C), Cerrillow 136 (Bi–Pb–Sn–In eutectic alloy, melting point of 58 °C), and Rose’s metal (Bi–Pb–Sn alloy, melting point of 98 °C), for low-temperature liquid metal batteries. All-liquid-metal batteries were built with an operating temperature of 100 °C (Figure 1a). Notably, In is relatively rare and mainly produced as a byproduct during the processing of other metals. Therefore, Rose’s metal, without the costly In element, shows great promise for stationary energy storage.

To gain more insights into the depressed melting points of multielement alloys, the charge density and atomic structure of eutectic Bi–Sn–In are investigated on the basis of ab initio molecular dynamics. Figure 1b shows the charge density contours of the Bi–Sn–In eutectic alloy, where an uneven distribution of electrons can be observed. It is noted that the charge density is the highest around Bi atoms, suggesting strong force field in the given atomic configuration. Moreover, the electron distribution near Sn and In atoms resembles that of the typical liquid metal of Hg, in which the electrons in the low-energy orbitals are strongly bound to atoms. Such a configuration is similar to the noble-gas molecules with extremely low melting points owing to the weak interaction. Compared with pure metals of Bi (271 °C), Sn (232 °C), and In (157 °C), the melting point of the ternary Bi–Sn–In alloy is remarkably reduced to 62 °C, which should be contributed by the altered charge density around metal atoms in the presence of adjacent heteroatoms, as revealed by ab initio molecular dynamics. The structure of the Bi–Sn–In alloy with contours of charge densities is provided in Figure 1c, and the relatively disordered arrangement of atoms should be beneficial to the lowered melting point as well. The isosurface charge density in Figure 1c correlates well with the contours of electrons in Figure 1b, indicating the weak interaction in Sn and In atoms.

During cell operation, good wetting of liquid metals on current collectors is the prerequisite to facilitate the charge transfer. Essentially, a strong interaction between liquids and substrates can cause the droplet to spread out over the solid surface, indicating a good wetting behavior. Herein, we tried different substrates as the potential current collector for liquid metal batteries. The contact angle between Bi–Sn–In and Al foil is presented in Figure 2a, and the liquid metal droplet balls up because of its large surface tension. Similar wetting behaviors can be observed using Cu (Figure 2b), stainless steel (SS, Figure 2e), and Ti (Figure 2f) as the current collector, suggesting unfavorable adhesive forces. In contrast,
the liquid metal wets Ga or Sn-coated current collectors very well, as shown in Figure 2c,d, suggesting the reinforced interaction between liquid metals and substrates. To shed light on the surface science, the density functional theory (DFT) modeling was constructed to unravel the interaction between Bi–Sn–In and different substrates. As shown in Table S1, it is evident that the wettability in Figure 2 correlates well with the magnitude of adsorption energy, and good wetting of the liquid metal on Ga and Sn should be induced by the strong interactions. To further visualize the adsorption effect, the electrostatic potential maps of the Bi–Sn–In liquid metal on different substrates were further simulated (Figure 3), where red regions with negative adsorption energy indicate strong binding while blue regions represent strong repulsion. The unit cell and crystal symmetry are different for different metals. Therefore, it shows different 2D symmetry when cleaving the surface. It is noted that the surface of Ga and Sn maintains more binding sites toward liquid metal atoms than other substrates. Accordingly, stronger interactions and anchoring effects are achieved to give rise to better wetting, which is in good agreement with the contact angles in Figure 2. Herein, the Sn-coated substrate was adopted as the current collector for liquid metal batteries considering its high abundance and low cost.

The redox characteristic of the Bi–Sn–In alloy is further studied using the galvanostatic discharge test. In view of the coexistence of three electroactive metals in the ternary alloy, the electrochemical test of pure metals was conducted for...
reference. Theoretical capacity of the Bi–Sn–In alloy can reach 502.9 mAh/g with the sodiation products of Na$_3$Bi, Na$_{13}$Sn$_4$, and Na$_2$In. As shown in Figure 4b, pure Bi exhibits two successive voltage plateaus at 0.7 and 0.5 V, respectively, corresponding to the reaction products of NaBi and Na$_3$Bi by alloying. As for Sn in Figure 4c, the first sodiation reaction occurs at 0.45 V, followed by the alloying reaction below 0.3 V. In comparison, a single voltage plateau was observed for In, suggesting the sole reaction between In and Na to get NaIn. In reference to the voltage profiles of pure metals, the discharge voltage profile of Bi–Sn–In can be divided into three regions (Figure 4a). The sodiation of Bi takes place first between 1 and 0.6 V, followed by multireactions involving the alloying of Bi, Sn, and In within 0.6 and 0.3 V. Notably, a minor potential hysteresis can be observed, which should be due to the interactions between multielements. To the end of discharge, the tiny trail in orange below 0.3 V should be ascribed to the further sodiation of Sn–Na alloys. The XRD patterns of Bi–Sn–In alloy and Bi, Sn, In metal are presented in Figure S1. It is noted that the XRD peaks of the discharged Bi, Sn, and In correlate well with reported works. Additionally, the XRD peaks of Bi–Sn–In alloy in a discharged state correlate with the peaks of discharged Bi, Sn, and In, as well. Meanwhile, some minor discrepancies may be ascribed to the changed environment of the multielement alloy in the presence of heterogeneous atoms. In deep discharge processes, the sodiated products of Bi–Sn–In typically maintain high melting points given the strong ionic character of the bonds. Consequently, the deep discharge operation of liquid metal batteries is always accompanied by a phase transformation with solid formation. Such a phase behavior is observed in other reported liquid metal batteries, and the solid phase can be converted back to liquids under charging, correlating to the self-healing nature of liquid metals. Herein, we narrowed the discharge cutoff potential to 0.6 V to alleviate the sluggish kinetics caused by solid formation under deep discharge. The composition evolution of the liquid metal corresponds to the sodiation of Bi to produce NaBi.

Conventional ester-based electrolytes will decompose at high temperatures, while ethers can withstand the harsh environments. After rational screening of different solvents and salts (Figure S2–S6), an electrolyte composed of 1 M NaI dissolved in tetraglyme was adopted given its thermal stability and compatibility. Figure 5a shows the voltage profiles of Bi–Sn–In versus Na at a current density of 4 mA/cm$^2$. Higher power performance can be expected by developing advanced electrolytes or making the electrolyte as thin as possible. The sole voltage plateau during the discharge process corresponds to the sodiation of Bi, correlating well with the first discharge region in black in Figure 4a. Thanks to the self-healing and dendrite-free feature of liquid metals, the voltage profiles over 40 cycles can almost overlap, showing application prospects for grid-scale energy storage. Although a trace amount of Sn as the coating layer of current collectors may be dissolved in the Bi–Sn–In alloy during battery cycling, the effect is negligible considering its mass ratio below 2% compared with the fusible alloy-based cathode. Meanwhile, good wetting is still maintained after cell cycling as presented in Figure S7, indicating the stable interface between liquid metals and current collectors. In contrast, when stainless steel was adopted, the liquid metal will ball up on the current collector with poor wetting (Figure S8). The voltage profiles in Figure S9 also show more obvious polarization with a larger overpotential. Figure Sb exhibits the cycling performance of Bi–Sn–In, and the Coulombic efficiency can be stabilized at ~100%, demonstrating the stable interface of both the anode and the cathode with negligible side reactions and self-discharge. In comparison, either molten salt-based electrolytes or ceramic-based solid electrolytes were adopted in reported high-temperature liquid metal batteries. Because of the high solubility of alkali metals, such as Na, in molten salts, especially at high temperatures, the Coulombic efficiency is imperfect with inevitable self-discharge. On the other hand, the costly and brittle solid electrolytes complicate the cell assembly and increase the capital cost of the energy systems. Herein, such a liquid metal battery with a low operating temperature is promising for practical applications. Although
the current density of the proof-of-concept battery cannot compete with high-temperature cells, enhanced power performance can be anticipated with the development of advanced electrolytes. In addition to the electrochemical performance, cost is also of vital importance to the practical application of energy-storage systems.\textsuperscript{45,46} Figure S10 compares the abundance of different chemical elements in the Earth’s crust. Despite the outstanding performance demonstrated, the low abundance and high market price of In limit the widespread implementation considering S1 wt % of In in the Bi–Sn–In alloy (Table S2). To decrease the capital cost, we can partially substitute In with Bi and Pb to make a Bi–Pb–Sn–In quaternary alloy called the Cerrolow 136 metal. Compared with the Bi–Sn–In-based ternary alloy which melts at 62 °C, the melting point of the Bi–Pb–Sn–In alloy is further decreased to 58 °C. The lowered melting point should be contributed by the optimized distribution of charge density around metal atoms in the presence of more heteroatoms.\textsuperscript{24} Figure S5c,d present the voltage profiles and cycling stability of the Bi–Pb–Sn–In liquid metal battery. It is evident that the superior cycling performance with 100% Coulombic efficiency confirms the self-healing capability of liquid metals. The depth of discharge of Na anodes is estimated to be ∼30%. The high plating/stripping efficiency of Na metal should be due to the dendrite-free and self-healable Na anode in the molten state. Figure S11 shows the stability of Bi–Pb–Sn–In versus Na over 100 cycles, and the ∼100% Coulombic efficiency can be maintained as well. To pursue an even lower-cost energy system further, we can completely substitute In with Pb/Bi to make a Bi–Pb–Sn ternary alloy. Although the melting point is lifted to 98 °C, it is still feasible to build a low-temperature liquid metal battery at 100 °C. Similar electrochemical performance with Bi–Sn–In and Bi–Pb–Sn–In is demonstrated, as given in Figure 5e, Sf, suggesting the universal characteristics of Bi-based fusible alloys toward high-performance liquid metal batteries.

The liquid metal battery prototype can be traced back to the 1960s, and the cell has witnessed great progress over the 60 years.\textsuperscript{6,47} Figure 6a compares the operating temperature of liquid metal batteries with various combinations of anodes and cathodes.\textsuperscript{21,25,48} It is noted that state-of-the-art liquid metal batteries typically rely on pure metals or binary alloys with high melting points. Accordingly, operating temperatures above 160 °C are usually needed to maintain the molten state of metallic electrodes. Although GaSn-based liquid metal battery can work at room temperature, the high cost of Ga restricts its practical applications for grid-scale energy storage. When using Na as the anode paired with Bi–Sn–In, Bi–Pb–Sn–In, or Bi–Pb–Sn alloys as cathodes, we build a liquid metal battery with an operating temperature as low as 100 °C. The Bi–Pb–Sn-based system is also promising in terms of material cost compared with Ga-based batteries. Therefore, it is speculated that the special maintenance related to sealing and corrosion of high-temperature liquid metal batteries can be circumvented, showing great potential for broad application beyond stationary energy storage. Moreover, the performance metrics of different fusible alloys in this work are also illustrated in terms of energy density, power density, cost, temperature, and toxicity in the radar chart (Figure 6b). Despite the relatively high melting point, the Bi–Pb–Sn alloy, possessing the highest amount of Bi without In contents, is appealing in the aspect of high energy density and low cost (Table S3). Therefore, the Bi–Pb–Sn-based liquid metal battery is promising for the next-generation energy technologies provided the health issue related to Pb should be taken into consideration.\textsuperscript{49} What’s more, given the lower cost of Pb and Sn than Bi, the demonstrated liquid metal battery based on Bi–Pb–Sn vs Na is also more cost-effective than the Na–Bi and Li–Bi systems in reported works.\textsuperscript{16} To conclude, we have developed a melting-point depression-enabled liquid metal battery at a low operating temperature of 100 °C with elucidated chemistry and attractive performance. The charge density, atomic structure, and interfacial chemistry of multielement alloys are unveiled by an integrated experimental and modeling study. It is inferred that the lowered melting point should be ascribed to the disordered arrangement of atoms and redistributed charge density in the presence of heteroatoms. Meanwhile, improved wettability of liquid metals is realized by enhancing the adhesion energy.\textsuperscript{50} Besides the lowered cost on cell maintenance, the low-temperature liquid metal battery also circumvents the issues related to brittle solid electrolytes or self-discharge using molten salt electrolytes in conventional high-temperature cells. Despite the volume expansion and shrink of liquid metals during cell operation, the electrode pulverization can be
avoided given the self-healing property of liquid metals, and attractive cycling stability is demonstrated for all three types of Bi-based fusible alloys with stabilized Coulombic efficiency. It is noted that reaction kinetics of liquid metals at intermediate temperatures may not compete with that at high temperatures. Currently the rate capability of the proof-of-concept battery is not yet comparable with reported systems because of the lower conductivity of organic electrolytes than high-temperature molten salts or solid electrolytes. Higher power performance can be expected provided advanced electrolytes will be developed. In this work, we studied the battery chemistry of ternary/quaternary fusible alloys, and the integrated experimental and computational approach is demonstrated to be a useful methodology to study the energy systems based on new battery chemistries. The cost-effective Bi–Pb–Sn ternary alloy represents a promising cathode alternative to design practical liquid metal batteries. The fusible alloys used herein represent a promising type of electroactive materials for the next-generation energy technologies. In view of the broad family of fusible alloys, the Bi-based alloys can be extended to other alternatives to build room-temperature all liquid metal batteries for more widespread applications beyond stationary energy storage.

ASSOCIATED CONTENT

Supporting Information
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Experimental methods and additional characterization results (PDF)

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