Solid-State Ionics in the 21st Century: Current Status and Future Prospects

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Abstract
The phenomenon of ion migration in solids forms the basis for a wide variety of electrochemical applications, ranging from power generators and chemical sensors to ionic switches. Solid-state ionics (SSI) is the field of research concerning ionic motions in solids and the materials properties associated with them. Owing to the ever-growing technological importance of electrochemical devices, together with the discoveries of various solids displaying superior ionic conductivity at relatively low temperatures, research activities in this field have grown rapidly since the 1960s, culminating in "nanoionics": the area of SSI concerned with nanometer-scale systems. This theme issue introduces key research issues that we believe are, and will remain, the main research topics in nanoionics and SSI during the 21st century. These include the application of cutting-edge experimental techniques, such as secondary ion mass spectroscopy and nuclear magnetic resonance, to investigate ionic diffusion in both bulk solids and at interfaces, as well as the use of atomic-scale modeling as a virtual probe of ionic conduction mechanisms and defect interactions. We highlight the effects of protonic conduction at the nanometer scale and how better control of interfaces can be employed to make secondary lithium batteries based on nanoionics principles. Finally, in addition to power generation and storage, the emergence of atomic switches based on cation diffusion shows great promise in developing next-generation transistors using SSI.

Introduction
Solid-state ionics (SSI) is an interdisciplinary research area where chemistry, physics, and materials science meet. This field of research concerns properties particularly associated with the motion of ions in solids. Although Takahashi\cite{1} first introduced the term SSI in the early 1970s, ionic transport phenomena in solids have been known for more than a century, going back to the early 1800s when Faraday discovered fluoride ion conduction in PbF\textsubscript{2}\textsuperscript{−}\textsuperscript{−}.\cite{2} In the past, SSI focused primarily on solid electrolytes, which are virtually pure ionic conductors in which the electric current is almost exclusively carried by charged ions. In fact, the discovery of solid electrolytes has engendered this new research field of SSI.

Some solid electrolytes have exhibited ionic conductivity as high as that of concentrated liquid electrolytes (~1 S cm\textsuperscript{−}\textsuperscript{−}). Consequently, solid electrolytes with such a high ionic conductivity have quickly attracted scientific and technological attention. One of the earliest applications employing solid electrolytes was the Nernst glower,\textsuperscript{3} a type of electrical lamp in which an oxide ion conductor (ZrO\textsubscript{2} containing 15 wt\% Y\textsubscript{2}O\textsubscript{3}) is used as a light source. More recently, polymer electrolytes (as exemplified by the perfluorosulfonate isomer Naion)\textsuperscript{4} have played a crucial role in the development of low-temperature oxidative fuel cells.\textsuperscript{5} The potential for practical use of solid electrolytes has been the driving force for rapid growth in research activities in SSI over the past several decades, since SSI provides the scientific underpinning for a wide variety of advanced electrochemical devices such as fuel cells, batteries, gas separation membranes, chemical sensors and, in the last few years, ionic switches. Mixed conduction, which arises not only from ion migration increased by modification of polarizability by the variation of electron density in the bandgap energy region but also via electrons or electron holes, plays a crucial role in electrochemical devices. Because the density of both ionic and electronic defects governs the kinetics of elemental charge transfer reactions, the research interest in this field has naturally been extended to mixed conductors.

From a microscopic standpoint, ion migration in solids is only possible because of the existence of point defects (i.e., vacancies and/or interstitials) in the solid bulk interior. The concentrations of point defects obey laws of mass action similar to those for dissociation equilibria in liquid electrolytes. In fact, the "defect chemistry" of solids is analogous to well-established solution chemistry in the liquid state. Therefore, the equilibrium thermodynamics of point defects and their diffusion kinetics in the bulk of solids, which have been the main focus of research in SSI during the past century, have been well-studied and understood.\textsuperscript{6−11}

While the basic concept of ion migration requires the presence of lattice imperfections, such as point defects, special interactions between mobile ions and the host framework are required for the attainment of even higher mobility, or “fast ion conduction.” The precise nature of the interactions between the framework and mobile ions is an unresolved issue; the “softness” or some local phonon dispersion may be a key to understanding this phenomenon, but the fact remains that we are currently still unable to systematically control the ionic mobility by modification of the atomic structure. The recent discovery of fast ion conductivity with low activation energy for the free oxyanion units in oxide ion and proton conductors (such as oxyapatite),\textsuperscript{12,13} CsHSO\textsubscript{4},\textsuperscript{14} LaNbO\textsubscript{3},\textsuperscript{15} suggests a new direction by which to control and modify the phonon-mobile ion interaction via the thermal activation of additional local rotational freedoms of such oxyanion units. This may allow for the realization of “super ionic conductors,” which have been proposed on the basis of the presence of special phonon/mobile ion interactions analogous to those in electrical superconductors. It is common for chemical stability, which is also of crucial importance in most applications, not to be sufficiently high in fast ion conductors.\textsuperscript{16} This is
probably due to a weak affinity between framework and mobile ions or, in other words, it is necessary for mobile ions to break their bonds with host ions before migration can occur, such as in the case of point defect–type ion conductors. In contrast to the crystallographic approach\textsuperscript{17–20} of the last century, the progress of a computational approach\textsuperscript{21–26} aided by the development of new hardware and resultant popularization in the materials science field, may help us to understand the long-standing mystery of super ionic transitions in the first and second order, as shown in Figure 1, with respect to the variation of effective mobile carriers and mobility, phonon-ion interaction, thermal activation, and stability of the phases.

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During the last decade, the behavior of point defects at interfaces (gas/solid, solid/solid, or both) and properties associated with this behavior have been the focus of extensive study in the field of SSI.\textsuperscript{27,28} Since the translational symmetry of solids is broken at interfaces, where relaxations occur in response to local changes in environment, the interfaces are expected to have atomic structures that differ inherently from the bulk structure. This fact suggests that the physical properties of the interface should differ markedly from those of the bulk.

In the nanometer regime, the interfaces of a crystalline solid are spaced extremely closely, such that their influences on the overall properties of the crystal are expected to become significant, if not decisive, and thus macroscopically measurable. Therefore, the distance between two neighboring interfaces becomes an additional control parameter in such a regime, determining the occurrence of “size effects.”\textsuperscript{27,29} Liang\textsuperscript{30} reported some of the first experimental evidence of an interfacially controlled materials property and demonstrated that Li\textsuperscript{+} conductivity measured in LiI was enhanced by nearly two orders of magnitude by dispersing fine Al\textsubscript{2}O\textsubscript{3} particles. It was concluded later that adsorption of Li\textsuperscript{+} on the Al\textsubscript{2}O\textsubscript{3} surface creates extra Li vacancies in the LiI:Al\textsubscript{2}O\textsubscript{3} composite, leading to the distinct enhancement in the conductivity in the material compared to the pure phase of LiI. More recently, Sata et al. reported enhanced fluoride ion conductivity in a planar heterostructure of CaF\textsubscript{2}/BaF\textsubscript{2} films.\textsuperscript{31} In this system, the fluoride ion conductivity increased by increasing the periodicity of the alternating films. Owing to thermodynamic reasons, near the contact between BaF\textsubscript{2} and CaF\textsubscript{2}, F\textsuperscript{−} ions in BaF\textsubscript{2} transfer to the adjacent CaF\textsubscript{2} across the contact, leaving their vacancies in BaF\textsubscript{2}. This leads to an increase in fluoride vacancy concentrations at the interfacial regions, which short-circuits the material. Such results triggered extensive studies on ionic conduction in various nanostructured ionic conductors. The research field of SSI concerning nanoscale systems is generally referred to as nanoionics.\textsuperscript{27}

The introduction of interfaces of nanoscale spacing brings about a new experimental degree of freedom for the field of SSI. Nano-sized structures with a high interfacial density can be sufficiently stable even in a non-equilibrium state to allow exploitation of their properties. This fact makes nanoionics one of the most promising new research areas of SSI in the 21st century. Nevertheless, the defect chemistry and related phenomena in nanostructured solids are still largely unstudied and thus deserve to be explored more extensively in the future. Accordingly, fundamental challenges involving measuring, understanding, and predicting ion behavior with respect to size variation must be addressed in order to ultimately engineer nanoionics devices.

In This Issue

This issue of MRS Bulletin provides an overview on a selection of research topics in SSI that currently attract researchers’ highest attention and provide a sound indication of future prospects. We have deliberately chosen not to confine these topics just to nanoionics, but, nevertheless, nanoionics is highly relevant to all six articles in this issue concerning key research issues.

As mentioned previously, detailed information about local variations in ion mobility in a solid will provide a unique opportunity to tailor the transport property of the materials. De Souza and Martin discuss diffusion kinetics in polycrystalline solid electrolytes, determined by using secondary ion mass spectroscopy techniques. Of particular interest in ionic diffusion in solid electrolytes is whether fast migration of ions along the grain boundaries is possible. Fast grain boundary transport in solid electrolytes would open up a second avenue for tailoring the transport properties of the solid electrolytes by optimizing the grain size.

Despite a good understanding of the general principles of ionic conduction in solids based on the macroscopic or classical point of view, a lack of in-depth knowledge about atomic-scale ion dynamics in these materials for the occurrence of diffusive migration has hindered the full development of fundamental predictive models of local conduction behavior. Information regarding the ion dynamics in solid ion conductors primarily comes from electrical conductivity measurements. However, such measurements reveal information on the average mobility of the ions undergoing long-range diffusion in the system with little direct microstructural understanding of the conduction mechanisms. An understanding of the more fundamental differences in atomic structure and defect distribution between interface and bulk and how they may control ionic conductivity is lacking. Heitjans and Wilkening describe how nuclear magnetic resonance spectroscopy can address these issues and provide such information.

Since the discovery of proton conduction in perovskite-structured oxides,\textsuperscript{32} there has been much interest in their use as solid-state electrolytes. The article by Norby is intended to review the recent research activities concerning protonic conduction in solids, including newly discovered protonic conduction phenomena in nanostructured solids at temperatures as low as room temperature. Both the scientific and technological significance of such non-traditional phenomena and their impact on the field of SSI will be

![Figure 1. The Arrhenius plot of typical ionic conductivity, \( \sigma \), of typical conductors that can be expressed by (1) a simple thermal activation process (Arrhenius behavior) of mobile ionic defects including protons; (2) glass transition or the second-order transition indicating non-Arrhenius behavior by the increase of disordered domains with rising temperature, observed in polyelectrolytes or beta-alumina; and (3) the first-order transition by sublattice melting in AgI and related compounds or a ferroelastic phase transition in superprotonic conductors, such as CsH\textsubscript{3}O\textsubscript{5}, GDC, gadolinium-doped ceria; SDC, samarium-doped ceria; YSZ, yttria-stabilized zirconia.](image-url)
addressed. One of the most important open questions about the chemical and phase stability of protonics materials, namely the acid/base interaction between host and protonic defects, is discussed in addition to the possible use of protons in semiconductors and photocatalytic reactions.

One of the most striking emerging topics in SSI is the application of ion migration for nonvolatile switching devices that can operate at room temperature. Significant impact was brought about by the broad recognition that ion migration can be sufficiently fast for gigahertz switching when the distance traveled is on the nanoscale. The article by Hasegawa et al. covers topics concerning atomic switches that employ mixed conducting sulfides and novel oxide systems with both metallic nanofilament and/or nonmetallic filament formation, with an emphasis on the importance of nanoscale ion migration. In the latter case, a foreign ion is donated from the electrode and migrates in a similar fashion to the electronic carrier donation and migration found in organic transistor materials.

The use of computational methodologies to understand the origin of fast ion migration in oxides and other compounds is rapidly developing due to the growth of computing power and tractable techniques for carrying out first-principles simulation, such as density functional theory. The article by Islam and Slater reviews such new advances in the emerging simulation and computational science of solid-state ionics, including recently discovered fast oxide ion and protonconducting oxides with unique and common crystal structural characteristics composed of free (i.e., not polymerized) oxycation tetrahedra that show fairly low activation energies for ion migration. A good example can be seen in oxide ion conductivity in anhydrous apatite-type oxides.

A Li+ ion battery is typically composed of a solid Li-intercalated graphite anode, a Li-intercalated oxide cathode with transition metals, and a Li+ ion conductor chosen from a wide variety of compounds. The development of an all-solid-state secondary Li-ion battery is key to realizing safe and high-power density energy storage systems, but it is well known that severe interfacial impedance prevails at the interface between the solid electrolyte and oxide cathode. Among various trials and approaches, a better control of interfaces based on nanomicroprinciples, by adding so-called buffer layers to prevent space-charge layer effects that develop at the interface, is becoming popular. In the article by Jamnik, these various approaches based on nanomicroprinciples to minimize the impedance at the interface will be reviewed and discussed.

Concluding Remarks

We conclude by recalling that the term solid-state ionics (SSI) is so named due to its similarity with solid-state electronics in the hope that it becomes equally familiar due to its technological impact on a whole engineering field. Such a dream may soon be realized in the form of nanoionics devices, such as the atomic switch and resistance random access memory, and by the utilization of nanoionics (i.e., the control of ion migration in nanoscopically structured materials). In the nanometer world, it is becoming increasingly clear that we can enjoy a freedom from the perennial problems of poor mobility and large effective mass of ions in micrometer/macro-scale applications. Ion migration in the nano to subnanoscale region can be key to making breakthroughs against Moore’s law in nanoelectronic, providing a basic function not only for nonvolatile memories, programmable integrated circuits, and memristors, but also for future possible applications to alternative complementary metal oxide semiconductor and photo-nanoionic devices using photoactivated migration. One also may be able to find further possible extensions of photo-nanoionics to photosynthesis and photocatalysis to utilize the local ion migration at the surface (e.g., heterophase-doped systems, as described in the article by Norby). In the case of a surface/interface with mobile ions that play an important role in the relaxation by the ionic defects, a soft compositional distribution with a very narrow and sharp potential profile may work as the charge separation site by the light irradiation, and one may hope to realize a novel photosynthesis device driven by ion migration.

To understand the nanoionics phenomena at a heterogeneous interface, the space-charge concept has been served as a basic feature of modification of both ionic and electronic defect concentrations, which contribute to the modulation of electrochemical kinetics governed by the population of both ionic and electronic species involved in the reaction. This is not a simple nano-size effect and can modify the macroscopic electrochemical kinetics governed by the interfacial properties, such as the interfacial impedance for ion migration. In addition to one-dimensional ionic diffusion at the interfaces, an understanding of higher dimensional ionic migration at the solid surface would widen our view of the nano-size effect in the future. The significance of interfacial tension, which contributes through the modulation of defect population by an elastic strain, could be one of many untapped frontiers that must be better understood in order to make use of interface properties in coming decades.

As we survey the prospect of SSI research of the 21st century, we would like to emphasize that the discovery of novel materials and novel ideas for applications always vitalize the scientific activity of solid-state ionics, and this effort must be vigorously pursued in the future. Recent advances notwithstanding, better ion conductivity is still required to achieve the unique benefits of the application of ionic materials, and it is hoped that this special issue contributes to stimulating further efforts in this direction.

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