The synthesis of inorganic materials by electrochemical methods has shaped the world we experience. From the design and production of microelectronics to the manufacture of the metal parts in construction, automobiles, and coinage, humanity has relied critically on inorganic materials electrosyntheses in one fashion or another for centuries. With continued innovations, electrosynthetic methods will only grow greater in societal importance and influence.

One basic area of opportunity for electrosyntheses is an expansion of the types of elements, compounds, and crystals attainable. Although elemental metallic materials are naturally suited for preparation electrochemically, electrosyntheses have the potential to be much more versatile in scope. Electrosyntheses of materials that are luminescent, chemically responsive, catalytic, or that exhibit quantized properties are necessary for the next generation of logic, sensing, and energy-conversion technologies. Through the development of richer palettes of electrolytes, electrochemical waveforms, and electrochemical cell/reactor designs, a variety of new forms of matter with such properties are accessible via electrosyntheses.

A second opportunity in inorganic electrosyntheses is the incorporation of 'single-entity' paradigms and concepts. For example, although electrosyntheses have been performed for several centuries almost exclusively for bulk materials, films, or ensembles of nanomaterials, electrochemistry is also naturally suited for isolated and independent synthetic reactions. The electrosyntheses of compounds by collisions between individual droplets containing electroactive precursors and poised electrodes will produce discrete reaction events that can be monitored, manipulated, and analyzed individually. Such methods could lead to entirely new alloy/intermetallic/oxide/chalcogenide/coordination polymer compositions and structures inaccessible by any other means because of differences in flux and concentration gradients at the microscopic scale. Similarly, the individual bubbles formed during solvent electrolysis side-reactions can be more than parasitic annoyances. These species could selectively and purposely shape the form factor (e.g. porosity, surface area) of electroformed species at the electrolyte/electrode interface, affording a new dimension of tunability.

A separate area where electrosynthetic methods continue to grow is the rapid, uniform, and conformal coating of textured surfaces. This capacity is one of electrosyntheses' greatest strengths, setting it apart from line-of-sight and (ultra) high-vacuum vapor phase deposition methods. High quality space-filling coatings by electrosyntheses are ideal for back-end-of-line processes in integrated circuit fabrication. As the density and design of circuits increase in both number and complexity, metal structures with even greater conformality are needed. Beyond just empirical recipes, electrosyntheses where the operative reaction mechanisms are understood are necessary to realize 'superconformal' films. Moreover, in combination with with redox-replacement strategies, the possible compositions of coatings are almost limitless.

A final area of great development in inorganic electrosyntheses is the ability to dictate the crystallinity of the product material. Although electrosyntheses routinely produce polycrystalline products, single crystal material growth is considerably more challenging but also generally more desirable, particularly with epitaxial control. The realization of electrosynthetic tactics that produce single-crystalline products will be aided by atomistic understanding of the initial stages of nucleation and crystal growth. The preparation of tailored single-crystalline products, especially at low temperatures and with the ability to be transferred to arbitrary types of substrates, will significantly expand the role of inorganic electrosyntheses in areas like the fabrication of green renewable energy technologies and flexible electronics.

This special issue of Accounts of Chemical Research describes a set of detailed works that comprise the latest innovations in these areas. Many of the leading research groups in inorganic electrosyntheses are contained herein, with their latest contributions to this vibrant field of science summarized and contextualized. These works suggest the breadth of products possible in inorganic electrosyntheses is only bounded by creativity and ingenuity. Collectively, this account on the *Electrosynthesis of Inorganic Materials* gives the reader a preview of the future of inorganic electrosynthesis for the next century and beyond.