Selective Ion Separations Using Shock Electrodialysis

by

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Agricultural development, extensive industrialization, and rapid growth of the global population have inadvertently been accompanied by environmental pollution. At the same time, the chemicals and energy industries face a number of difficult challenges in the selective extraction of high-value ionic species from dilute aqueous solutions. Important examples include the selective removal of trace pollutants, such as toxic heavy metal ions, from industrial effluents; the fractionation of chemically and physically similar elements, such as lanthanides for catalytic processes; the extreme deionization of wastewaters, such as radioactive process water from nuclear power plants; and the recovery of lithium compounds and valuable metals for applications in mining and electronic waste recycling. These emerging trends have motivated the search for new principles and methods for improved ion separations. Electrochemical methods in particular have attractive features such as compact size, modularity, chemical selectivity, broad applicability, and reduced generation of secondary waste.

In this thesis, we investigate the emerging electrokinetic approach known as "shock electrodialysis" (shock ED) and its use in selective ion separations. Although the principles of deionization by shock ED have been established in previous work, the possibility of selective ion separations has only recently been discovered, and this capability is explored in depth in this thesis. The first major thrust is an extensive and comprehensive review of electrochemical methods for water purification, ion separations, and energy conversion. The review begins with an overview of conventional electrochemical methods, which drive chemical or physical transformations via Faradaic reactions at electrodes, and proceeds to a detailed examination of the two primary mechanisms by which contaminants are separated in nondestructive electrochemical processes, namely electrokinetics and electrosorption, with special attention given to emerging methods such as shock ED. The second major thrust is the design of processes and operating conditions to demonstrate the broad applicability of shock ED for selective ion removal from contaminated water. We developed several design concepts to control the selective separation of cations, anions, and small, charged hydrocarbons based on electric charge. The third major thrust is the examination of new types of materials in shock ED, including ceramics, clays, and ion exchange resins, several of which enable operation under extreme conditions (e.g., high temperature, high radiation, chemically harsh or reactive contaminants). This study led to the development of shock ion extraction (shock IX), which is a new, hybrid process that combines shock ED and ion exchange and enables greater ion removal and selectivity, and for longer periods of time, compared to the use of either shock ED or ion exchange alone.

From a fundamental perspective, the novel electrokinetic mechanisms explored in this thesis are shown to have broader implications in deionization, water purification, and metals refining. For the field of chemical engineering, this work demonstrates shock-based methods as an energy-efficient and sustainable route to process intensification, and it paves their way for practical implementation in industry.

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