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Review article

# Surface and interface sciences of Li-ion batteries -Research progress in electrode–electrolyte interface-

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## ABSTRACT

The application potential of Li-ion batteries is growing as demand increases in different fields at various stages in energy systems, in addition to their conventional role as power sources for portable devices. In particular, applications in electric vehicles and renewable energy storage are increasing for Li-ion batteries. For these applications, improvements in battery performance are necessary. The Li-ion battery produces and stores electric power from the electrochemical redox reactions between the electrode materials. The interface between the electrodes and electrolyte strongly affects the battery performance because the charge transfer causing the electrode redox reaction begins at this interface. Understanding of the surface structure, electronic structure, and chemical reactions at the electrode–electrolyte interface is necessary to improve battery performance. However, the interface is located between the electrode and electrolyte materials, hindering the experimental analysis of the interface; thus, the physical properties and chemical processes have remained poorly understood until recently. Investigations of the physical properties and chemical processes at the interface have been performed using advanced surface science techniques. In this review, current knowledge and future research prospects regarding the electrode–electrolyte interface are described for the further development of Li-ion batteries.

## 1. Introduction

Concerns regarding environmental pollution and the depletion of energy sources in our life are increasing. The most realistic and feasible strategy to solve these problems is the enhancement of the efficiency of energy use. Energy usage at a specific time and place

*Abbreviations:* ABF, annular bright field; AFM, atomic force microscopy; AM, amplitude modulation; EC, ethylene carbonate; CVD, chemical vapor deposition; DEC, diethyl carbonate; DFT-MD, density functional theory-molecular dynamics; DMC, dimethyl carbonate; DME, dimethoxyethane; DMSO, dimethyl sulfoxide; DOS, density of states; DR-XAS, depth resolved X-ray absorption spectroscopy; EDX, energy dispersive X-ray; EELS, electron energy loss spectroscopy; EIS, electrochemical impedance spectroscopy; EMC, ethyl methyl carbonate; ESD, electrostatic spray deposition; FEC, fluoroethylene carbonate; FM, frequency modulation; GC, gas chromatography; HAADF, high-angle annular dark-field; HAX-PES, hard X-ray photoelectron spectroscopy; HOPG, highly oriented pyrolytic graphite; HREELS, high resolution electron energy loss spectroscopy; IR, infrared; LEED, low energy electron diffraction; LLT,  $\text{La}_{0.55}\text{L}_{10.35}\text{TiO}_3$ ; MD, molecular dynamics; MS, mass spectroscopy; NR, neutron reflectivity; PE, polyethylene; PEO, polyethylene oxide; PES, photoelectron spectroscopy; PLD, pulsed laser deposition; PM-FTIR, polarization-modulation Fourier-transform infrared; PP, polypropylene; PVdF, polyvinylidene difluoride; RF, radio frequency; SEI, solid electrolyte interphase; SEIRA, surface enhanced infrared absorption; SEM, scanning electron microscopy; SERS, surface enhanced Raman scattering; SFG, sum frequency generation; SOC, state of charge; SLD, scattering length density; STEM, scanning transmission electron microscopy; STM, scanning tunneling microscopy; STO,  $\text{SrTiO}_3$ ; STS, scanning tunneling spectroscopy; TEM, transmission electron microscopy; TERS, tip enhanced Raman scattering; TRF-XAS, total-reflection fluorescence X-ray absorption spectroscopy; UHV, ultra high vacuum; VC, vinylene carbonate; WKB, Wentzel-Kramers-Brillouin; XPS, X-ray photoelectron spectroscopy; XRD, X-ray diffraction; XRR, X-ray reflectivity

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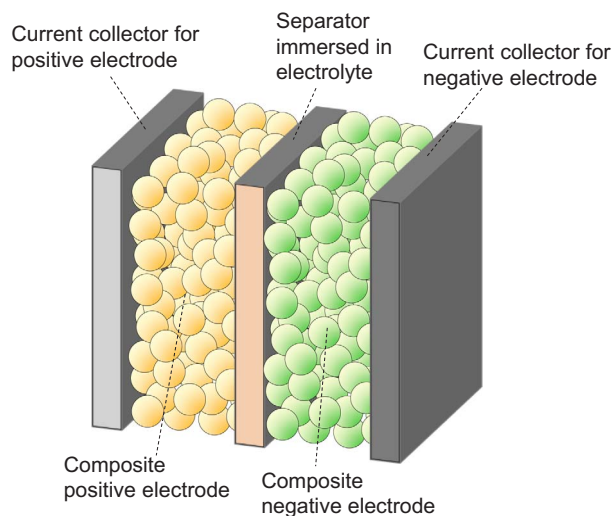


Fig. 1. Schematic model of typical structure of an industrial Li-ion battery.

by generating electricity using a rechargeable battery is one suitable method for improving efficiency. Currently, Li-ion batteries are the most common type of rechargeable batteries, with widespread use in portable electronic devices. Recently, the application of Li-ion batteries has expanded to electric vehicles, factories, and renewable energy storage [1–4]. To meet the demands of these new applications, Li-ion batteries require improvements in capacity, cycling, and high-rate performance. To improve battery performance, the interface between the electrodes and electrolyte must be both thoroughly understood and controllable. In this review, current knowledge and future research prospects regarding the electrode–electrolyte interface in Li-ion batteries are summarized.

The first Pb–acid rechargeable battery was invented in 1859 by Gaston Plante; the development of nickel–cadmium and nickel–hydrogen batteries followed, and finally led to rechargeable batteries using Li [1–4]. The high specific capacity (3860 A·h/g) of Li suggested its use as an electrode material; Li–TiS<sub>2</sub> and Li–MoS<sub>2</sub> batteries are examples of Li-ion battery systems. However, the low recharging behaviors and safety issues of Li negative electrodes induced the replacement of Li-negative electrodes with intercalated materials. In lithiated carbon (typically graphite), the charge–discharge reaction in an appropriate organic electrolyte forms a stable surface film known as the solid electrolyte interphase (SEI) on the electrode [5]. The applicability of LiCoO<sub>2</sub> in positive electrode material was discovered by Goodenough *et al.* in 1980 [6]. Current Li-ion batteries use graphite negative electrodes and lithiated-metal layered compounds, typically LiCoO<sub>2</sub>, as positive electrodes, in which the Li insertion and extraction processes occur during the charge and discharge reactions, respectively. In 1991, Li-ion batteries were commercialized by SONY for use in small electric devices because they offered high energy densities [7]. At present, Li-ion batteries are indispensable power sources for portable electronic devices in modern life [1–4,9].

Modern industrial Li-ion batteries are typically composed of the following components (Fig. 1):

- A. A current collector for the negative electrode
- B. A composite negative electrode
- C. A separator to prevent direct contact between the negative and positive electrodes immersed in an organic electrolyte
- D. A composite positive electrode
- E. A current collector for the positive electrode

Typically, Cu is used for A. B is prepared by mixing the active material with a binder. Carbon materials and F-containing resins like polyvinylidene difluoride (PVdF) are typically used as the active and binder materials, respectively. When active materials with low electron conductivity are used, the conductive material, i.e. acetylene black is mixed with the composite electrode. For C, polyethylene (PE) or polypropylene (PP) microporous membranes are used as separators. The separator is immersed by the electrolyte, typically a Li salt such as LiPF<sub>6</sub>, is dissolved in a carbonate-based organic solvent, often a mixture of ethylene carbonate (EC) and dimethyl carbonate (DMC), diethyl carbonate (DEC), or ethyl methyl carbonate (EMC). Small amounts of additives such as vinylene carbonate (VC) are also dissolved in the electrolyte to improve the battery performance. For D, a mixture of the active material, binder, and conductive materials is prepared (Fig. 2) [8]. Layered metal oxides like LiCoO<sub>2</sub> and PVdF are typical active and binder materials, respectively. To improve the electron conductivity, carbon materials like acetylene black are used as the conductive material. For E, Al foil is typically used, with the composite positive electrode pasted onto the metallic current collector. During the charge–discharge reaction in a Li-ion battery, Li ions and electrons move by diffusion between the active materials in the negative and positive electrodes.

During the charging and discharging of a Li-ion battery, Li ions move between the positive and negative electrodes. Here, it is assumed that Li ions initially occupy the positive electrode (Fig. 3). To begin the discharging reaction, a lower electrochemical potential is applied to the positive electrode. To compensate for the difference in electrochemical potential between the positive and

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