



# Emerging applications of spark plasma sintering in all solid-state lithium-ion batteries and beyond

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## HIGHLIGHTS

- Comprehensive review of spark plasma sintering (SPS) for solid-state batteries.
- SPS enables intimate grain-grain boundary in inorganic solid-state electrolytes.
- SPS allows good electrode-electrolyte interfaces in solid-state Li-ion batteries.
- Improved solid-solid interfaces leads to enhanced solid-state battery performance.
- Highlights of SPS in fabrication of solid-state Li-ion, Li-S, and Na-ion batteries.

## ARTICLE INFO

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## ABSTRACT

Solid-state batteries have received increasing attention due to their high safety aspect and high energy and power densities. However, the development of solid-state batteries is hindered by inferior solid-solid interfaces between the solid-state electrolyte and electrode, which cause high interfacial resistance, reduced Li-ion and electron transfer rate, and limited battery performance. Recently, spark plasma sintering (SPS) is emerging as a promising technique for fabricating solid-state electrolyte and electrode pellets with clean and intimate solid-solid interfaces. During the SPS process, the unique reaction mechanism through the combination of current, pressure and high heating rate allow the formation of desirable solid-solid interfaces between active material particles. Herein, this work focuses on the overview of the application of SPS for fabricating solid-state electrolyte and electrode in all solid-state Li-ion batteries, and beyond, such as solid-state Li-S and Na-ion batteries. The correlations among SPS parameters, interfacial resistance, and electrochemical properties of solid-state electrolytes and electrodes are discussed for different material systems. In the end, we point out future opportunities and challenges associated with SPS application in the hot area of solid-state batteries. It is expected that this timely review will stimulate more fundamental and applied research in the development of solid-state batteries by SPS.

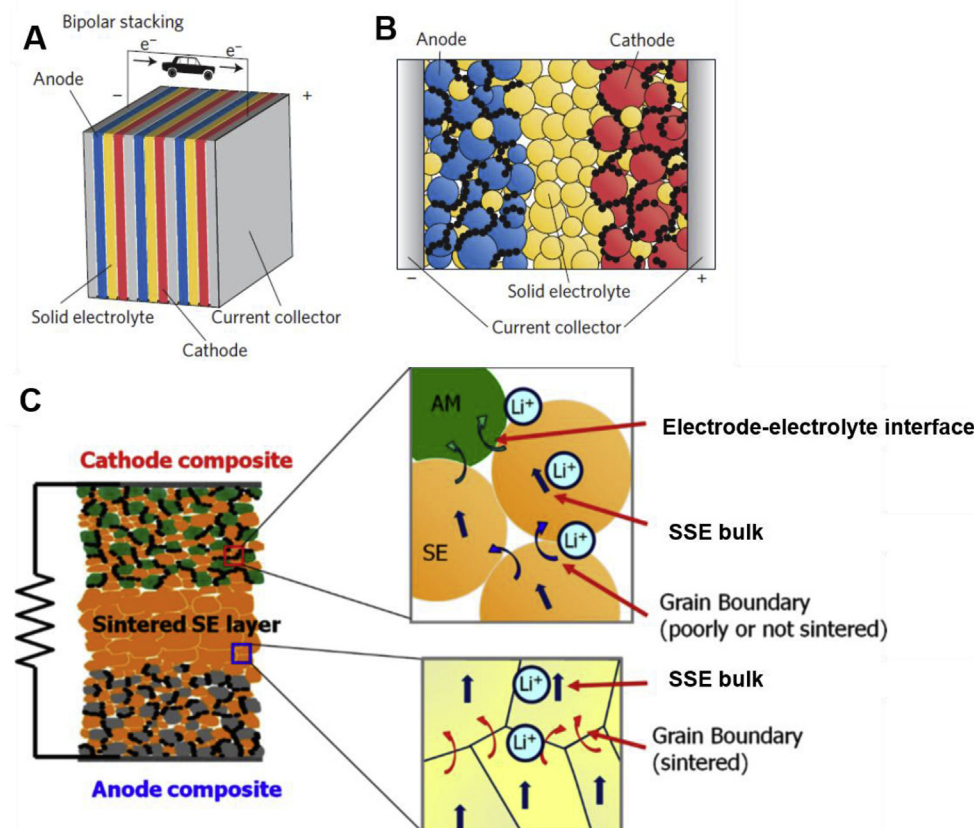
## 1. Introduction

Lithium-ion (Li-ion) batteries have been used as energy storage systems in a wide range of applications, such as portable electronics and electric vehicles, due to their relatively high energy and power density among available technologies [1–3]. However, traditional Li-ion batteries based on liquid organic electrolytes have difficulty in meeting demand from automotive industries for batteries with higher energy density (350–500 Wh kg<sup>-1</sup>), faster charge/discharge rate, better safety, and lower cost. In particular, traditional Li-ion batteries suffer from potential safety risks, such as thermal runaway and explosion, due to the highly volatility and flammability nature of liquid organic

electrolytes employed [1–3]. Therefore, all solid-state Li-ion batteries (ASSLIBs) have regained the position of paramount research interest in a hope to overcome the above problems in traditional Li-ion batteries [4–11]. In ASSLIBs, solid-state electrolytes (SSEs) are the key component used to replace liquid electrolytes used in traditional Li-ion batteries [12–16]. The typical structure of an ASSLIB consists of cathode, SSEs, anode, and current collectors, as seen in Fig. 1. In general, there are two types of configurations, bipolar and pellet-type, for ASSLIBs, as shown in Fig. 1a and b, respectively [14,17]. In a bipolar configuration (Fig. 1a), the electrode are coated on the two sides of the current collectors, which could reduce the weight and volume of the battery package and increase the energy density of batteries [14,17]. In a

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**Fig. 1.** (A) A bipolar configuration of ASSLIBs, (B) a pellet-type configuration of ASSLIBs, and (C) structure of an all solid-state batteries and two types of solid-solid interfaces: grain-grain interface in the SSEs and electrode-electrolyte interface in the electrode composites. Reprinted with permission from Ref. [17] Copyright 2016 Springer Nature; Reprinted with permission from Ref. [18]. Copyright 2015 American Chemical Society.

pellet-type configuration (Fig. 1b), one layer of each anode, SSEs, and cathode is stacked in a sandwich structure, providing lower energy density than bipolar one due to the thin layers of the electrode and thick layer of SSEs [14,17]. Current research development in ASSLIBs focuses on the pellet-type configuration, because it is easier to fabricate than bipolar one and suitable for fundamental studies in the electrode. In the ASSLIBs, SSEs function both as ion-conducting pathways in the cathode and anode composites, and as separator between the two electrodes, and therefore play a critical role in ASSLIBs. For the working principle of an ASSLIB, during the discharge process, Li-ions exist the lattice structure of the anode, pass through the anode-SSE interfaces, enter the SSE bulk and separator, and then move across the SSE/cathode interfaces, and finally diffuse into the lattice structure of anode, while electrons transfer to the cathode via the external circuit. During the charge process, the Li-ions and electrons flow in the reversed direction. As we can see from the discharge/charge process, the efficient Li-ion diffusion process is critical for the overall battery performance. The use of SSEs not only eliminates the safety risks resulted from liquid electrolytes, but also opens the door to adopting Li metal as the anode in the battery system, thus substantially increasing the gravimetric and volumetric energy densities of ASSLIB cells [19,20]. Moreover, in ASSLIBs, no organic liquid electrolyte, electrolyte salt, additional separator, or binder is required, thus potentially simplifying the assembly and packaging processes and reducing the fabrication cost [4]. Furthermore, the development of SSEs and safe Li metal anode paves the pathways towards several other promising new battery technologies, such as lithium-sulfur (Li-S), lithium-oxygen (Li-O<sub>2</sub>) batteries [21–29].

In order to develop ASSLIBs, one of the critical challenges is the inferior solid-solid interfaces in the electrolyte and electrode, which could result in high interfacial resistance and poor cycle performance [4,5,23]. In ASSLIBs, there are main two types of solid-solid interfaces in the electrode and electrolyte that could become rate-limiting parameters for the electrochemical reactions [18]. The first one is the grain-

grain interfaces in SSEs, as schematically shown in Fig. 1c. The role of SSEs is to separate the anode from the cathode, and transport Li ions between them during charge/discharge process. Thus, high total ionic conductivity (bulk + grain-boundary) is a prerequisite for SSEs to be deployed in ASSLIBs. During the past decades, great progresses have been made to improve the bulk ionic conductivity of inorganic SSEs via different approaches, such as cation doping [12,16,30,31]. However, the low grain-boundary conductivity, resulted from high interfacial resistance at the poor grain-grain interfaces, becomes one major bottleneck for the total ionic conductivity of inorganic SSEs [18,23,31]. The other type of solid-solid interfaces is the electrode-electrolyte interfaces between the electrode materials and SSEs, as well as the interface between electrode and carbon additives (Fig. 1c). The main problem at the electrode-electrolyte interfaces includes small contact area between electrolyte and electrode and poor connection of the active material with the carbon additives in the electrode [4,5]. The poor electrode-electrolyte contacts could result in high charge transfer resistance at the interfaces, and thus low rate capability, efficiency, and cycle stability of ASSLIBs. Since the SSE and carbon additives serve as the diffusion paths for Li ions and electrons out of/to the electrode materials, good electrode-electrolyte interfaces will ensure fast Li-ion and electron transport for efficient electrochemical reactions to occur in ASSLIBs. In practice, the electrode-electrolyte interface situation could become more complicated, due to several other problems involved at the interfaces, such as the volume change in the electrode materials, the electrochemical stability between the electrode and electrolyte, etc. [4,5,32]. Therefore, it is imperative to achieve good solid-solid contacts, at both grain-grain and electrode-electrolyte interfaces, in order to reduce the interfacial resistance and achieve longevity as well as high energy and power densities in ASSLIBs.

The solid-solid interfaces are strongly determined by the electrode and electrolyte sintering techniques. Several sintering techniques, such as cold press and hot press, have been employed to fabricate electrode

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