



Parametric study of dry coating process of electrode particle with model material of sulfide solid electrolytes for all-solid-state battery



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ABSTRACT

We have reported that a dry coating is a very promising process to produce a suitable composite particle for all-solid-state lithium-ion secondary batteries. We here present a parametric study of this dry coating process for further optimization and improvement. In the dry coating process, electrode particles were coated with solid electrolyte (SE) particles. $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ (NCM) was used as a host particle, while sodium sulfate (Na_2SO_4) was selected as a model material of sulfide SEs and used as a guest particle. A dry impact-blending process known as “Hybridizer” was used. Effects of critical process parameters including tip speed of rotor, processing time, and initial weight fraction of host and guest particles were investigated. Under insufficient impact force NCM particles were not fully covered with Na_2SO_4 particles, while under a sufficient impact force the discrete coating of Na_2SO_4 particles on the surface of NCM particles were observed. With an increase in the mechanical input energy per unit mass of Na_2SO_4 , plastic deformation and coalescence of Na_2SO_4 particles were promoted, resulting in the continuous coating. With further increase in the mechanical input energy, breakage of the NCM particles was caused. The continuous coating exhibited a great advantage over the discrete coating for formation of the solid-solid interfacial contacts between NCM and Na_2SO_4 in the compressed pellet. Finally, the optimal processing conditions, where NCM particles are able to be coated by the continuous layer of much less amount of Na_2SO_4 (= 10 wt%) without breakage of NCM particles, were determined.

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1. Introduction

A new next generation secondary battery for large-scale power storage devices has been strongly required for hybrid electric vehicles (HEVs) and electric vehicles (EVs), renewable energy productions, smart grid applications, and so on [1,2]. Lithium-ion secondary batteries (LIBs), which are widely used in portable electronic devices, are considered as a potential candidate for the large-scale secondary battery. However, LIBs have a serious safety concern for application to the large-scale battery, because the organic liquid electrolytes used in the current LIBs are flammable. As a promising candidate of next generation secondary battery, all-solid-state lithium-ion secondary batteries (ASS-LIBs) have attracted much attention [3–5]. In the ASS-LIBs, inorganic solid electrolytes (SEs) are used instead of the organic liquid electrolytes. SEs are non-flammable materials and stable under higher voltage and broad temperature range. Due to these features, the ASS-LIBs are highly expected to be a next generation secondary battery with both high safety and high energy density.

For development of the ASS-LIBs, there are two major issues: (1) synthesis of new inorganic materials for SEs, and (2) development of processing technologies to produce composite electrodes. The lithium-

ion conductivity of typical SEs was lower than that of organic liquid electrolytes [4]. To date, material scientists have synthesized new inorganic materials with higher lithium-ion conductivity. Among the candidate materials, sulfide SEs have been extensively investigated for the ASS-LIBs [3,4,6–10]. Recently, new sulfide SE which exhibits equivalent lithium-ion conductivity to organic liquid electrolytes has been synthesized [10]. However, high performance of the ASS-LIBs cannot be accomplished only by synthesizing new materials for SEs. In the ASS-LIBs, transfer of lithium ions during charging and discharging only occurs at the interfacial contact surface between an electrode particle and SE. This requires well-designed composite particles and electrodes in which solid-solid interfacial contacts between electrode particles and SEs for the lithium-ion conduction pathway is formed [3,4]. Moreover, breakage of the electrode particles, that can lead to negative impact on the battery performance of the ASS-LIBs, should be avoided. Therefore, development of processing technologies to construct the solid-solid interfacial contacts in the composite particles and electrodes without breakage of the electrode particles is also important issue.

Many studies on the processing technologies to produce the ideal composite particles and electrodes have been reported. Some simple powder mixing techniques such as the use of smaller size of SEs [11] and wet mixing of electrode particles and SE particles [12] have been studied. These studies indicate that performance of the ASS-LIBs can be improved even by these simple powder handling techniques. A

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planetary ball milling [13,14] has often been used for mixing of electrode particles and SE particles. To achieve better composite particles, agglomerates of fine electrode and SE particles should be disintegrated sufficiently without breakage of the electrode particles. However, no one has succeeded in such an ideal mixing, suggesting that the planetary ball milling is less suitable for the processing technology to produce the composite particles and electrodes. To produce better composite particles and electrodes, it is ideal that the primary electrode particles are individually coated with SEs without breakage of the electrode particles. Such an ideal coating has been examined by means of a pulsed laser deposition (PLD) [15,16]. In this technology, SE layer can be deposited on surface of the electrode particles, resulting in coating of primary electrode particles with the SE. The authors demonstrated a significant improvement in the battery performance using the coated particles. However, the PLD has disadvantages of low throughput and much expensive running cost due to significant low formation rate of the coating layer. Thus, the PLD is very expensive and far from practical use in an industrial process [4]. Apart from the powder handling equipment, hot pressing at temperatures above the glass transition temperatures of SEs [17] have also been attempted. However, the heating treatment can reduce the battery performance.

In our previous study [18], we proposed to apply a dry coating process and reported that individual primary electrode particles (host particles) are able to be successfully coated with a model material of sulfide SEs (guest particles). In a compacted pellet (i.e., mimic of composite electrode) prepared from the coated particles, the solid-solid interfacial contacts between electrode particles and the model material of SEs are well formed, suggesting that the dry coating process is very effective to produce composite particles and electrodes with the ideal structure for the ASS-LIBs. However, effects of many processing conditions on the coated particles have not been investigated, and there is much room for further improvement and optimization of the process.

We here present a parametric study of the dry coating process. $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ (NCM) which is a typical cathode active material was used as a host particle. Sodium sulfate (Na_2SO_4) was selected as a model material of sulfide solid electrolytes and used as a guest particle. The dry impact-blending device was used as a dry coating device. Effects of critical process parameters including tip speed of rotor, processing time, and initial weight fraction of host and guest particles were systematically investigated. Through this investigation, mechanism of the dry coating in this study was discussed. Finally, the optimal processing conditions were determined.

2. Experimental

2.1. Equipment

A dry impact-blending process (Hybridizer NHS-0, Nara Machinery Co., Ltd.) was used as a dry coating device (Fig. 1). This device mainly consists of chamber (inside diameter of 125 mm, depth of 49 mm, and effective volume of $470 \times 10^3 \text{ mm}^3$), rotor with six blades (diameter of 118 mm), and circulation pipe. The dry coating was conducted as follows. A pre-mixed powder mixture of host and guest particles was fed into the chamber. The rotor was then rotated and the particles circulate in the device. During the circulation, the particles are subjected to high impact forces due to particle-to-wall and particle-to-particle collisions. As a result, agglomerates of the particles are disintegrated and surface of the host particles are covered with guest particles, resulting in the dry coating. After a predetermined processing time, the discharge valve is opened, and coated particles are collected.

2.2. Powder samples

$\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ (NCM) (NCM-03, Toda Kogyo Corp.) and sodium sulfate (Na_2SO_4) (JIS special grade, Wako Pure Chemical Industries, Ltd.) were used as powder samples. Fig. 2 shows the volume based

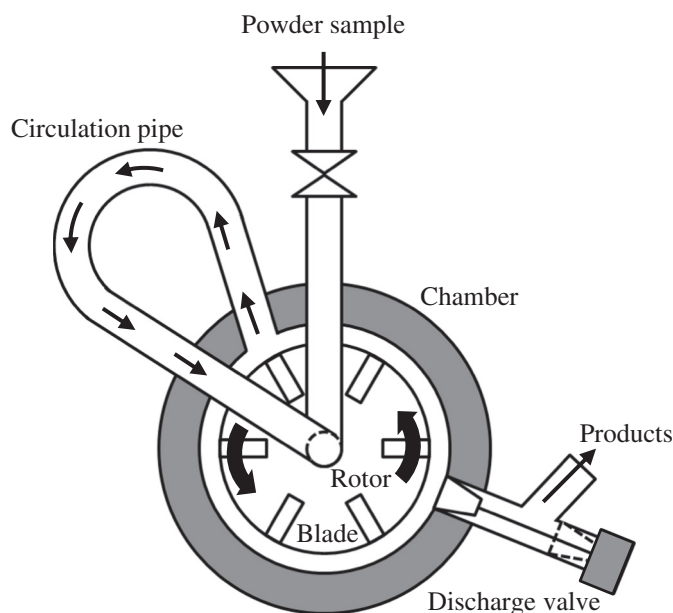


Fig. 1. Schematic of dry impact-blending device (Hybridizer).

particle size distributions and SEM images of the powder samples. NCM is a typical positive-electrode active material for LIBs. Volume based median diameter of the NCM was $5.4 \mu\text{m}$. NCM is relatively rigid, brittle, and electrically conductive material, as compared to Na_2SO_4 . NCM was used as a host particle in the dry coating. Na_2SO_4 was selected as a model material of sulfide SEs [18], because sufficient amount of sulfide SEs to conduct our experiments is commercially unavailable. For selecting a model material of sulfide SEs, we considered that following properties are important: (i) nature of the room-temperature pressure sintering; (ii) mechanical properties such as stiffness and elastoplasticity. It has been reported that sulfide SEs exhibit the unique densification property so called “room-temperature pressure sintering [16]”: when powder of a sulfide SE is compressed, the sulfide SE particles are able to be easily deformed and coalesced even at a room temperature. This results in a compressed pellet in which the original SE particles are continuously coalesced, and void spaces and particle boundaries are almost vanished. In this study, the model material of sulfide SEs was selected from a few inorganic materials which exhibit the room-temperature pressure sintering. We then measured the stiffness and elastoplasticity of the candidate materials by means of an indentation test [18]. As a result, Na_2SO_4 showed the room-temperature pressure sintering as well as similar stiffness and elastoplasticity with a sulfide SE [18]. Consequently, Na_2SO_4 was selected and used in this study as a model material of sulfide SEs. Na_2SO_4 is relatively soft, ductile, and non-conductive material, as compared to NCM. Na_2SO_4 was used as a guest particle of the dry coating. The as-received powder of Na_2SO_4 was preliminarily milled by a multi-ring-type wet grinding system (MICROS-0, Nara Machinery Co., Ltd.). Volume based median diameter of the Na_2SO_4 after the wet milling was $0.95 \mu\text{m}$.

2.3. Experimental procedure and conditions

NCM and Na_2SO_4 were pre-mixed by mortar-and-pestle mixing for 30 min. This pre-mixture was then processed by the dry impact-blending device. Table 1 shows processing conditions of the dry coating. Effects of the tip speed of rotor, processing time, and initial weight fraction of NCM and Na_2SO_4 particles were investigated. Total weight of NCM and Na_2SO_4 were 10.0 g in all experimental runs. The dry coating experiment was conducted one time at each Run No., except for the Run No. 11. We confirmed that there is reproducibility in the dry coating experiment and there is no need to consider the batch deviation.

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