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Adhesion of tin-iron-soda-silicate anode glass to solid electrolyte ceramics by laser processing

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Abstract

Combining the viscous flow of glass melt and a local heating process by laser, a tin-iron-soda-silicate $55SnO$-$15Na_2O$-yFe_2O_3-$(30-y)SiO_2$ (y=0-9) anode paste was deposited on the solid electrolyte to develop all-solid-state sodium battery. A micro melt pool was formed in a laser condition up to the power of 3 W and scanning speeds of 100 mm/s. As a result, the anode layer became densified and adhered to the solid electrolyte without any external load to press. Compared to conventional sintering, the bonding is completed faster, and undesirable crystallization at the interface is avoided. This method provides a breakthrough in forming hetero junction structures between battery materials with a mismatch in processing temperatures.

1. Introduction

All-solid-state batteries (ASSBs) are expected to be next-generation batteries because of their lower cost and higher safety. However, the high interface resistance between the electrode and solid electrolyte is one of the barriers to the practical use of ASSBs. In general, oxide materials must be sintered at high temperatures to integrate the electrode and solid electrolyte[1)]. However, it has been reported that a cathode made of glass ceramics can form a junction interface with a solid electrolyte by laser irradiation[2)3)]. On the other hand, an anode material for ASSBs and the junction of anode and solid electrolyte by laser irradiation have yet to be developed. We reported tin-iron-soda-silicate glass anode in the previous study[4)]. We propose new technique to form hetero junction structure compose of anode glass-ceramics and solid electrolyte for ASSBs.

2. Experimental

The tin iron soda-silicate glasses, $55SnO$-$15Na_2O$-yFe_2O_3-$(30-y)SiO_2$ (y = 0-9) (yFe-SNS) was prepared by planetary ball milling a SnO and sodium iron disilicate glasses, $33.3Na_2O$-xFe_2O_3-$(66.7-x)SiO_2$ (x = 0-20). 6.75Fe-SNS powder was mixed with polyimide binder in a weight ratio of 85:15 to synthesize the anode paste. The anode paste was applied on a solid electrolyte substrate of about 10 μm thickness by a screen printing method. Then, a laser beam of 1064 nm wavelength was irradiated in two dimensions at a laser power of 3 W and a scanning speed of 100 mm/s or higher to form a junction interface with the solid electrolyte substrate (Fig.1).

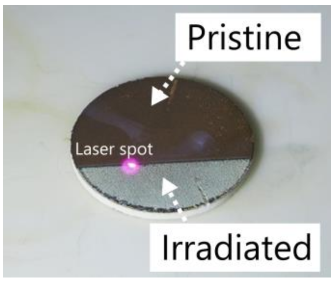

Fig.1 Laser irradiation of 6.75Fe-SNS on solid electrolyte.

3. Results and discussion

3.1 Comparison of sintering and laser irradiation processes

6.75Fe-SNS paste was deposited on the solid electrolyte, the conventional sintering and laser irradiation processes were performed on the sample, and the interface bonding between the anode electrode and the solid electrolyte was characterized. The conventional heat treatment did not densify the anode and bond it to the solid electrolyte, laser irradiation at a laser power of 3 W and a scanning speed of 100 mm/s resulted in densification of the anode and bonding with the solid electrolyte.

3.2 Morphological evaluation of bonding interface

Fig.2 shows the bonded sample's cross-sectional FE-SEM image and EDX mapping. The morphology of the cross-section consists of three regions: the region of the solid electrolyte substrate where grain boundaries are observed, the region where solid electrolyte components are densified, and the region of the densified anode layer. Each component is not mixed in each region, and unlike the conventional bonding interface formed by heat treatment, no reaction occurs between the materials. It was found that laser irradiation affected not only the anode layer but also the densification of the solid electrolyte.

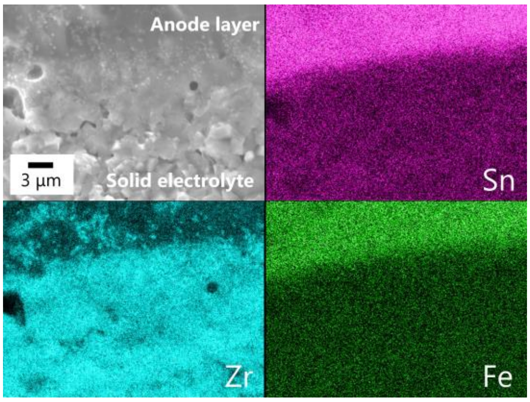

Fig.2 FE-SEM image and EDX mapping of cross section.

4. Conclusions

We formed an anode/solid electrolyte heterojunction structure by laser irradiation to practically use all-solid-state sodium batteries. Tin-iron-soda-silicate glass anode was deposited on the solid electrolyte. Laser irradiation at a laser power of 3 W and a scanning speed of 100 mm/s resulted in densification of the anode and bonding with the solid electrolyte. This discovery is the first report on preparing an integrated sample by forming a bonding interface between the anode active material and the solid electrolyte using the laser irradiation process shown in this study. It is expected to be used in all-solid-state batteries.

Acknowledgments

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