Preparation and evaluation of non-sintering type all-solid-state battery using garnet-type solid electrolyte

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All-solid-state battery expected to have high safety and energy density are being studied as one of next generation batteries. All-solid-state battery has sulfide-type and oxide-type, the former tends to be plastically deformed, therefore it can be densified by pressurization and interface formation among particles. However, it can release toxic H_2S gas when exposed to moisuture in the atmosphere. On the other hand, although the oxide-type is excellent in stability in the atmosphere, it is hard and hardly deformed, so it is necessary to densify and use a method such as sintering or vapor deposition to form interface among the particles. However, both methods are not suitable for increasing size. Even in the case of oxide, if it can be densified only by pressurization like the sulfide, it is easy to upsize the battery.

In this study, we investigated the electrolyte material which can be densified without sintering, although it is an oxide type, by adding a material which is plastically deformable. As the oxide solid electrolyte, a garnet-type structure ($Li_7La_3Zr_2O_{12}$[1]) is advantage of a high ion conductivity and a wide potential window, which is currently spotlighted, and LiI which is low ion conductivity was selected as an additive material because this is tend to occur plastic deformation which is effective for densification.

LLZ-Mg,Sr($Li_{7-2x+y}Mg_xLa_{3-y}Sr_yZr_2O_{12}$) was used as the LLZ-based solid electrolyte. The ionic conductivity of the LLZ-Mg,Sr sintered body was 1.4×10^{-3}S/cm@25˚C[2]. LLZ-Mg,Sr+LiI composite powder (50:50 (vol%)) was prepared by a dry planetary ball mill and a molded body was fabricated by pressing. The total conductivity of the LLZ-Mg,Sr+LiI molded body was 2.0×10^{-5}S/cm@25˚C(Fig.1). The ionic conductivities of the molded bodies of LLZ-Mg,Sr powder and LiI powder are on the order of 10^{-8}S/cm and 10^{-7}S/cm respectively, and moreover, the LLZ-Mg,Sr+LiI was found that the conductivity was higher than that of a single substance. We are currently preparing and evaluating non-sintering type all-solid-state battery using this composite solid electrolyte, and we plan to report on these results including on the day.

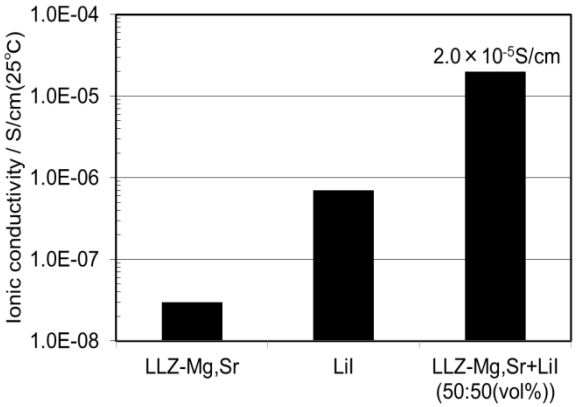

Fig.1 Ionic conductivity of molded bodies LLZ-Mg,Sr, LiI and LLZ-Mg,Sr+LiI

References:

[1] R.Murugan et al., *Angew. Chem. Int. Ed*., 46, 7778 (2007)
[2] Y.Takeuchi et al., 57th Battery symposium in Japan,1G23(2016)