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A Sinterless Garnet $Li_7La_3Zr_2O_{12}$ Thick Film as a Basis of All-Solid-State Li-Ion Battery

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A thick film of garnet type cubic $Li_7La_3Zr_2O_{12}$ (c-LLZO) with 1 wt% Al was prepared from a paste comprising c-LLZO nanoparticles synthesizd via a mechanochemically assisted solid-state route [1] and Li-ion conducting polymeric binder (Li-PAA) with varying Li : PAA molar ratio. Structural, chemical and electrochemical properties of the thick film was examined by using various analytical tools, among others, ^{6}Li MAS NMR. The Li ion conductivity peaked at the molar ratio, Li : PAA = 3 : 1, with the value of $8.15 \cdot 10^{-5}$ S $\cdot$ cm^{-1}, which is half of that of the sintered pellet [1]. The chemical shift of the main ^{6}Li NMR peak is significantly lower from Li-PAA than that of LLZO, as shown in Fig. 1, indicating strong exchange of Li species between Li-PAA and c-LLZO. The observed upfield shift by Li-PAA is presumably associated with the partial hydration of Li^+. The mixture of LLZO and Li-PAA exhibits its chemical shift just between these two values, indicating the Li^+ exchange between those of LLZO and Li-PAA. Interpretation of these changes in the ^{6}Li NMR spectra will further be elucidated from the viewpoints of atomic distance and coordination number by virtue of detailed X-ray diffractograms and vibration spectra, which are now on going. Some preliminary results of the electrochemical properties of the thick film by using a half cell indicates potentiality of the present thick film to apply to the all solid Li-ion batteries.

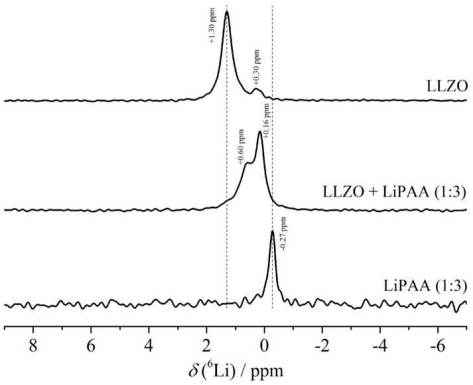

Figure 1: ^{6}Li NMR spectra of LLZO power, LLZO-Li-PAA thick filme and Li-PAA binder.

[1] A Novel Low-Temperature Solid-State Route for Nanostructured Cubic Garnet $Li_7La_3Zr_2O_{12}$ and its Application to Li-Ion Battery, P. Jeevan Kumar, K. Nishimura, M. Senna, A. Düvel, P. Heitjans, T. Kawaguchi, N. Sakamoto, N. Wakiya, H. Suzuki, RSC Adv. 6 (2016) 62656.

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