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Novel Composite Electyltes Using Single-Ion Conducting Fumed Silicas

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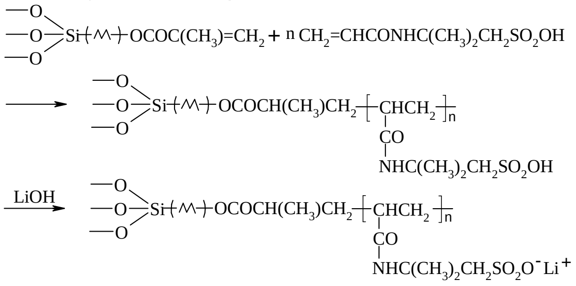
Since polyethylene oxide (PEO) electrolytes were first reported by Wright et al.,[1] polymer electrolytes have been considered an important research field for lithium battery application. The conductivity of most conventional electrolytes is based on the mobility of both cations and anions. The migration of anions results in a polarization potential, which consequently decreases ionic transport. Attempts to design single-ion conducting polymer electrolytes by covalently anchoring anionic moieties to the polymer chains have, despite ingenuity, met with limited success either in terms of conductivity or electrochemical stability.[2] One possible approach to solve this issue, proposed in our laboratory, is to disperse nanometer-size fumed silica with tailored surface characteristics into a liquid electrolyte and form a continuous network that mechanically supports the solution and yields a gel-like consistency.[3,4] A single-ion conductor is obtainable by attaching moieties capable of complexing with Li^+ cations onto the silica surface. In this communication, we present a novel single-ion conducting fumed silica [R711-poly(lithium 2-acrylamido-2-methyl-1-propanesulfonate), R711-pLiAMPS] in which lithium 2-acrylamido-2-methyl-1-propanesulfonate (LiAMPS) is anchored onto the surface of acrylate–functionized fumed silica (R711, Degussa) using a free-radical reaction followed by ion exchange.



High-transference number composite electrolytes were prepared by dispersing R711-pLiAMPS into oligomeric polyethylene glycol dimethyl ether (PEGdm), where lithium imide salt (lithium bis(trifluoromethylsulfonyl)imide, LiTFSI, 3M) may be added to assist ionic transport.

Figure 1 shows the effect of Li^+ surface concentration (γ) on the room-temperature conductivity for R711-pLiAMPS + PEGdm composite electrolytes (Li:O mole ratio = 1:100). Figure 2 shows the effect of Li:O mole ratio on the room-temperature conductivity of R711-pLiAMPS (γ = 4.2 nm^{-2}) + PEGdm composite electrolytes. From Figures 1 and 2, it is seen that a maximum room-temperature conductivity of 4.5 $\mu S\ cm^{-1}$ is obtained at a surface Li^+ content of 4.2 nm^{-2} and a Li:O mole ratio of 1:100 (15.8 wt % filler). The Li^+ transference numbers of R711-pLiAMPS (γ= 4.2 nm^{-2}) + PEGdm composite electrolytes were measured to be 0.59 and 0.78 at a Li:O mole ratio of 1:100 and 1:20 (48.5 wt % filler), respectively.

LiTFSI salt was also added to provide additional (and more mobile) charge carriers. It is found that adding LiTFSI to the samples enhances room-temperature conductivity by more than 2 orders of magnitude while still maintaining a relatively high-Li^+ transference number.

Acknowledgements

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References

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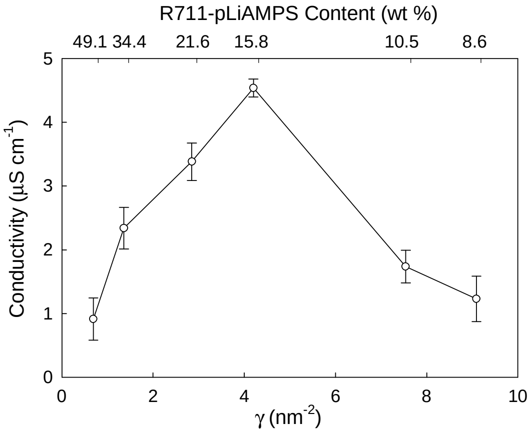

Figure 1. Room-temperature conductivity as a function of Li^+ surface concentration γ for R711-pLiAMPS + PEGdm (250) composite electrolytes (Li:O = 1:100).

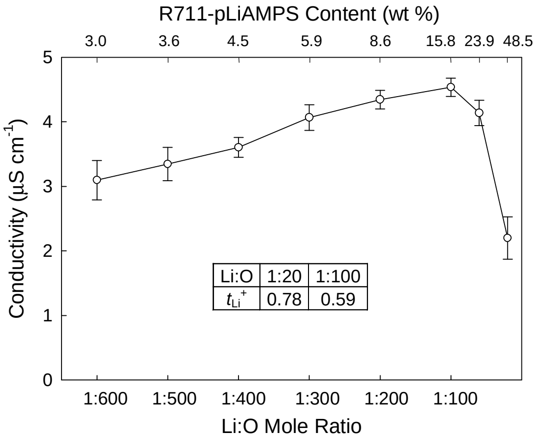

Figure 2. Room-temperature conductivity as a function of Li:O mole ratio for R711-pLiAMPS (γ = 4.2 nm^{-2}) + PEGdm (250) composite electrolytes. Inset table reports lithium transference number for two compositions.

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