New Proton Conductive Composites with Multi-Heterointerface Structures

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Ultra-thin multilayer of solid electrolytes can realize the artificial interfaces, where the distortion of the lattice and the space charge layers effectively enhance the ionic conductivity [1]. Approaches taken to increase ionic conductivity by utilizing heterointerfaces have been reported in recent years, and the preparation procedures of such interfaces include depositing superlattice structures by a pulsed laser deposition [2], and dispersing ceramic nanopowders [3]. In this study, new proton conducting composites with *Multi-Heterointerface Structures* were prepared. First, phenylsilsesquioxane ($PhSiO_{3/2}$) microparticles for the core of multilayers were prepared by the sol-gel method. Spherical $PhSiO_{3/2}$ particles in the micrometer scale were obtained by varying the molar ratios of ethanol, water, hydrochloric acid, ammonia and phenyltriethoxysilane in the starting sols. Second, multilayers of positively charged poly(diallyldimethylammonium chloride) (PDDA) and negatively charged Nafion® or phosphotungstic acid (PWA) were alternately deposited on the negative-charged $PhSiO_{3/2}$ core particles via layer-by-layer assembly technique (Fig.1). The formations of each layer were confirmed from changes in ζ-potential of the particles. It was found that multilayers consisting of PDDA and Nafion® or PWA were successfully deposited on the $PhSiO_{3/2}$ cores by these processes. Sheet-like electrolytes with good thermal and hydrolytic stability can be obtained from the core-shell particles by pressing. For example, proton conductivity of the electrolyte prepared using PWA-deposited particles was about 10^{-4} S/cm at 60 °C under 90% RH, the value of which was 5 orders of magnitude higher than that of the electrolyte prepared using unmodified $PhSiO_{3/2}$ particles. The composite sheet with multi-heterointerface structures is a promising electrolyte for fuel cells operating at medium temperatures.

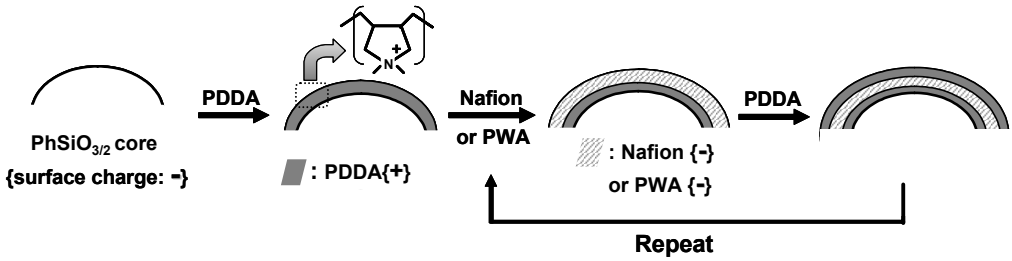

Fig. 1. Schematic illustration of the surface modification of core particles with Nafion or PWA via layer-by-layer assembly technique.

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