

Tape/Freeze-cast PVDF-HFP/Silica Composite Membranes for Battery Separators

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Abstract:

Advantageous characteristics of separators for Na-ion batteries such as directionally aligned pores for shorter transport paths, high porosity, and hydrophilicity for greater electrolyte uptake, can be achieved by tape/freeze casting of polymeric solutions with ceramic fillers as reinforcements. In this study, the introduction of silica particles from the sol-gel reaction of tetraethoxysilane (TEOS) into poly (vinylidene fluoride-hexafluoropropylene) (PVDF-HFP) membranes is achieved by a co-solvent method or by a subsequent infiltration of the reinforcement. The effects of TEOS on the microstructure, crystal structure, hydrophilicity and mechanical properties of tape/freeze-cast membranes have been investigated for both co-solvent method and infiltration process. The tape/freeze-cast PVDF-HFP membranes fabricated with dimethyl sulfoxide (DMSO) as the solvent and liquid nitrogen as the coolant exhibit directionally aligned and dendritic pores, while a hierarchical pore morphology with spherical pores on the aligned pore walls is found in membranes fabricated with TEOS as a co-solvent. Although silica is incorporated into membranes via both the co-solvent method and the infiltration process, as confirmed by FTIR and EDX, the infiltration process is found to be more effective in introducing silica. With the introduction of silica, the composite PVDF-HFP/SiO₂ membranes exhibit similar porosities but higher tensile strengths and a noticeable increase in electrolyte uptake and decrease in contact angle due to enhanced hydrophilicity compared to its polymer counterpart.

Keywords: Tape/freeze casting, PVDF-HFP, membranes, batteries, separators