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Highly dispersible graphene oxide nanoflakes in pseudo-gel-polymer porous separators for boosting ion transportation

Jin Il Kim [a], Jae Sang Cho [b], Dong Hwan Wang [b, **], Jong Hyeok Park [a, *]

[a] Department of Chemical and Biomolecular Engineering, Yonsei University, 50 Yonsei-ro, Seodaemun-gu, Seoul, 120-749, Republic of Korea
[b] School of Integrative Engineering, Chung-Ang University, 84 Heukseok-ro, Dongjak-gu, Seoul, 156-756, Republic of Korea

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A B S T R A C T

Gel-type polymer electrolytes have received considerable attention due to the battery explosion issue associated with volatile liquid-electrolyte-based lithium ion batteries (LIBs). However, the high ionic conductivity of gel-type polymer electrolytes originates from polymer swelling by the liquid electrolyte, and these materials inevitably have poor mechanical strength during device deformation. Here, we report structural gel-type polymer separators with highly porous and uniform morphology arising from the phase inversion of PVdF-HFP polymers with highly dispersible nanoscale graphene oxide nanoflake (GON). Via simple γ-ray irradiation of conventional graphene oxide solution, large 2D particles were cut into small 2D particles with a narrow size distribution, which in turn resulted in a dramatic change in solution transparency and particle dispersity. γ-ray-irradiated graphene oxide nanoflakes (γ-GON) with high dispersity are located inside the porous PVdF-HFP skeleton, inducing additional micron-sized pores of ~8 μm in the composite membranes. The modified porous film showed both gel-polymer electrolyte-like (uptake of 1.7 times more liquid electrolyte than conventional polyethylene separator) and polymer separator-like behavior (maintenance of original porous structure after soaked with liquid electrolyte). As a result, this pseudo-gel-polymer separator with a tailored pore structure has uniform ion flux and enhanced interfacial properties with electrodes, contributing superior battery performance.

1. Introduction

With the growing attention given to energy conversion and storage technology, lithium ion batteries (LIBs), which are common energy storage devices, have been researched and developed over decades [1–3]. Because of the high operating voltage and specific energy density of LIBs, these devices have been used in various industrial applications, such as IT devices, electronic vehicles (EVs), and energy storage systems (ESSs) [4–7]. Until now, research regarding LIBs has focused on electrode material design (cathodes and anodes) to satisfy the demand for a powerful specific energy density [8–11]. However, safety issues such as thermal runaway and battery explosions have been reported, arising from precarious separators with weak thermal stability, so more researches on reinforced separators is necessary to solve this safety drawback [12,13].

To enhance the thermal and mechanical properties of separators, thermally robust inorganic materials have been adopted for manufacturing inorganic/organic composite separators. Studies on the preparation of inorganic/organic separators via solution coating and casting methods have reported the embedding of inorganic particles into organic substrates [14,15]. Such inorganic-material coating processes on polyethylene (PE) separators enable enhanced thermal properties, but it is difficult to control the morphology of the packed inorganic layer to regulate ion flux and interfacial stability between the cathode and anode [16,17]. Atomic layer deposition and chemical vapor deposition, which have merits in the generation of coating layers on the substrate surface or nanoscale pores inside, are also methods for producing thermally sturdy separators [18,19]. However, a smaller pore size can hinder ion flux, especially at high current densities, leading to inefficient charge/discharge processes despite the introduction of a relatively high-cost technique. Above these, there are some reports about an additive using graphene oxide to fabricate thermally and mechanically stable separator [20–22]. Graphene oxide has an effect

* Corresponding author.
** Corresponding author.
E-mail addresses: king0401@cau.ac.kr (D.H. Wang), lutts@yonsei.ac.kr (J.H. Park).

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