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Development of intermediate temperature sodium nickel chloride rechargeable batteries using conventional polymer sealing technologies

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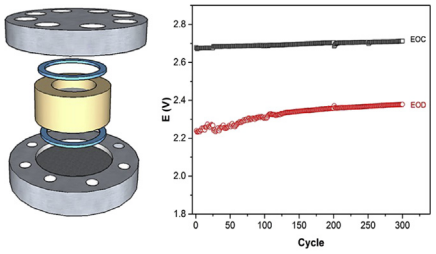
HIGHLIGHTS

- Conventional polymers were adopted as sealing materials for Na-$NiCl_2$ batteries.
- Selected polymers showed excellent compatibility with sodium and melts at 190 °C.
- Batteries with polymer seals showed stable performances over 300 cycles.

GRAPHICAL ABSTRACT



$2Na + NiCl_2 \leftrightarrow 2NaCl + Ni$ at 190 °C
(charge state) (Discharge state)

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ABSTRACT

Developing advanced and reliable electrical energy storage systems is critical to fulfill global energy demands and stimulate the growth of renewable energy resources. Sodium metal halide batteries have been under serious consideration as a low cost alternative energy storage device for stationary energy storage systems. Yet, there are number of challenges to overcome for the successful market penetration, such as high operating temperature and hermetic sealing of batteries that trigger an expensive manufacturing process. Here we demonstrate simple, economical and practical sealing technologies for Na-$NiCl_2$ batteries operated at an intermediate temperature of 190 °C. Conventional polymers are implemented in planar Na-$NiCl_2$ batteries after a prescreening test, and their excellent compatibilities and durability are demonstrated by a stable performance of Na-$NiCl_2$ battery for more than 300 cycles. The sealing methods developed in this work will be highly beneficial and feasible for prolonging battery cycle life and reducing manufacturing cost for Na-based batteries at elevated temperatures (<200 °C).
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1. Introduction

Na-based batteries have gained recognitions for next-generation stationary energy storage systems due to highly abundant and low cost Na resources and its suitable reduction potential (−2.7 V vs. a standard hydrogen electrode) [1–5]. Among all Na-based batteries under development, Na-metal halide (Na-MH) batteries, in which the cathode materials are composed of transition metal halides such as $NiCl_2$, $FeCl_2$, or mixed Ni-$FeCl_2$, have become one of the best candidates to satisfy the increasing energy demand for a wide range of energy storage applications [6–10].

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The essential parts of Na-MH systems are a molten sodium anode, a β″-alumina solid-state electrolyte (BASE), a secondary electrolyte and a metal-halide cathode. A Na-MH battery can be assembled in a fully discharged state using sodium chloride (NaCl) and metal powders without handling extremely pyrophoric metallic sodium and hygroscopic metal halides. The BASE, a sodium-ion-conducting ceramic, prevents crossover and battery self-discharge (100% coulombic efficiency) for a broad range of operational conditions. Furthermore, the use of an inorganic secondary electrolyte, sodium aluminum tetrachloride ($NaAlCl_4$), provides high Na^+ ion conductivity and superior safety (low fire hazard level) [7,11–14]. Despite these attractive features, Na-MH batteries have not successfully achieved further market penetration because of challenges including complicated cell architecture, high operating temperature and degradation of battery performance over cycles [15–19].

Typically, a conventional tubular type Na-MH battery is operated at high temperatures above 280 °C to obtain acceptable electrochemical performances. Our recent work on a planar type Na-MH battery demonstrated that remarkable electrochemical performances can be achieved even at an intermediate temperature (IT) of 190 °C [20–22]. The overall electrochemical performance of the IT Na-MH battery exhibits a stable cycle life of over 1000 cycles with an excellent capacity retention. The high energy density and superior cycle stability are attributed to the slower particle growth of cathode materials (NaCl and Ni) at 190 °C compared to that at 280 °C [21]. Moreover, lowering the operating temperature of a Na-MH battery from 280 °C to 190 °C opens great opportunities to adopt cost-effective sealing technologies for the cell architecture. For instance, the high operating temperature requires complicated and high cost sealing methods (such as glass seals, thermal compression bonding and electron beam welding), which are critical for Na-MH batteries to achieve airtight and corrosion-free seals against a molten sodium anode and a halide-based secondary electrolyte. In contrast, the lower operating temperature of 190 °C could allow conventional polymers to be used as sealing materials in Na-MH batteries.

In this study, we have evaluated and optimized various conventional polymer materials, which are listed in Fig. 1, as candidates for sealing materials in planar type Na-$NiCl_2$ batteries. A prescreening test was performed to quickly select sealing materials among those high temperature polymers for the cathode and the anode of the battery operated at 190 °C. The selected polymers were implemented in Na-$NiCl_2$ cells, and their compatibility was investigated by monitoring the electrochemical performance during the cell cycling. It has been shown that the polymer-sealed cells could be cycled more than 300 cycles with stable cell performance. Use of the conventional polymers demonstrated in this work can be a cost-effective, reliable and practical solution for Na-MH battery technologies, and provides a great opportunity for developing low cost planar sodium battery technologies.

2. Experimental

2.1. Polymer prescreening tests

Prescreening tests were performed in a glove box on eight different types of polymer films: ultra-high-molecular-weight polyethylene (UHMW-PE), polyvinylidene fluoride (PVDF), polytetrafluoroethylene (PTFE), fluorinated ethylene propylene (FEP), perfluoroalkoxy alkane (PFA), polyether ether ketone (PEEK), polyimide (Kapton), and ultra-temperature polyetherimide (Ultem PEI), which were purchased from McMaster-Carr. Na ($T_m = 97.8$ °C) and $NaAlCl_4$ (secondary electrolyte, $T_m = 157$ °C) in glass vials were placed on a 200 °C hot plate and the polymers were soaked in the molten Na and $NaAlCl_4$ for 5 min. PTFE, FEP, and PFA are immersed

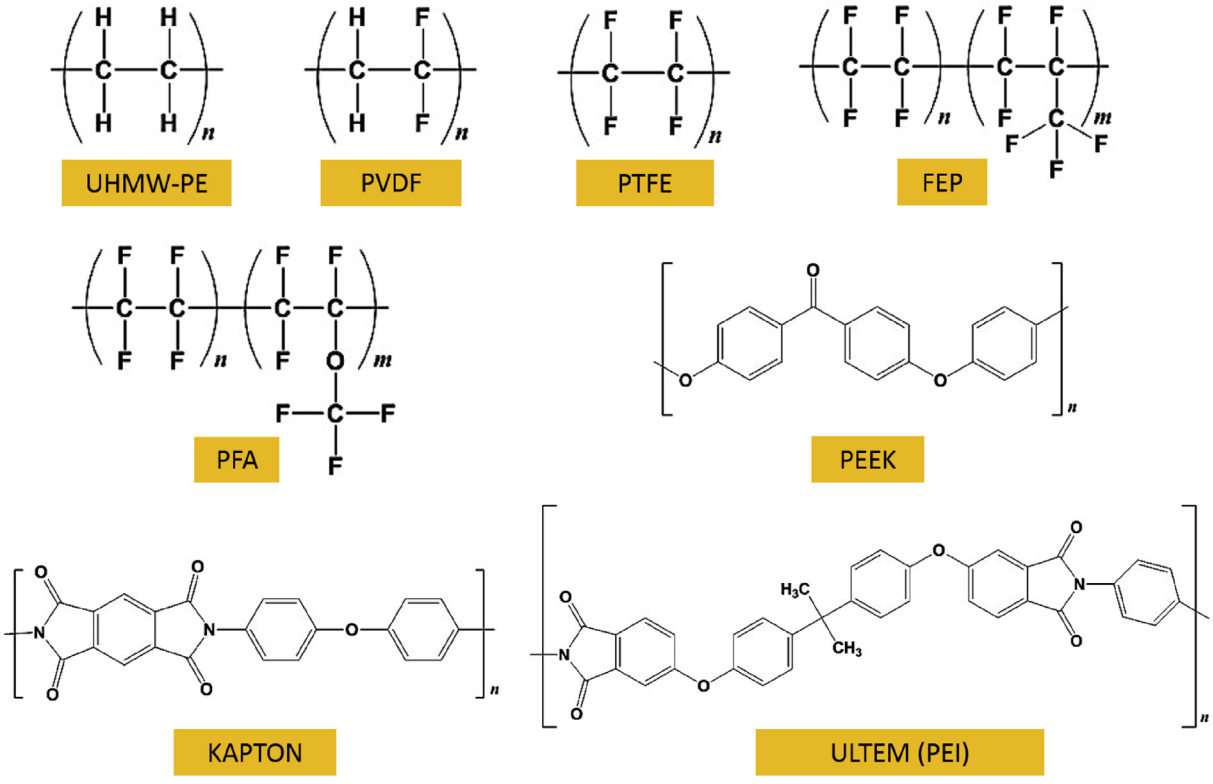

Fig. 1. Schematic structure and composition of polymer candidates as sealing materials.