Sodium ion rechargeable battery

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a Design concept for the alkaline aqueous battery. b Cycling performance for three cells at 1 C. c Cycling performance for NMF//NTP cell with Ni/C coating at 0.5 C at a low temperature of -30 ?C. d Long-term cycling stability for NMF//NTP full cell with Ni/C coating at 10 C, 1 C = 118 mA g-1, based on NMF.

Prepared NMF, NTP and commercial Ni/C powders were subjected to X-ray diffraction (XRD, Supplementary Figs. 1-3), evidencing good crystallinity for applications in batteries. The impact of salt concentrations in electrolytes on HER was established via in-situ differential electrochemical mass spectrometry (DEMS) in NaClO4 electrolytes with selected salt concentrations. Findings confirm that, without forming a reliable SEI, increasing electrolyte concentration does not change the onset potential for HER (Supplementary Fig. 4).

a Cycling performance for NMF//NTP pouch cell at a current density of 500 mA g-1. b Cycling performance for NMF//NTP pouch cell at 300 mA g-1. c Digital photograph of a cut pouch cell to power a fan in water. d Digital photograph of a cut pouch cell to power a humidity clock in water. e Comparison of lifespan and energy density with reported ASIBs. f Comparison of present work with commercial batteries (as quantified in Supplementary Table 4).

a Schematic for Ni/C coating for mitigating structural instability because of Mn dissolution in NMF cathode. Charge/discharge curves for NMF//NTP cells in (b) Neutral electrolyte (c) Alkaline electrolyte and (d) Alkaline electrolyte with Ni/C coating. e Operando Raman spectra for Ni/C coated NMF cathode cycled in alkaline electrolyte. f STEM-EDS spectra taken from NMF electrodes following cycling in neutral, alkaline electrolyte and alkaline electrolyte with Ni/C coating. g STEM line scan for cycled NMF cathodes with Ni/C coating. Inset shows Ni mapping for NMF.

The introduction of Ni was also confirmed via STEM-EDS mapping (Fig. 4f). There is a new peak belonging to Ni element in the spectrum. Additionally, the EDS line scan spectra for a single NMF particle confirm that Ni atoms are introduced into the edge of particles to suppress the dissolution of inner Mn atoms (Fig. 4g). The STEM-energy-dispersive spectroscopy (STEM-EDS) mappings for NMF cathode with Ni/C coating following 1st, 5th and 20th cycles (Supplementary Table 6 and Supplementary Fig. 21) evidence that the content of Ni in NMF particles is stable after the first cycle, confirming that the introduction of Ni into NMF cathode reaches an equilibrium in the first cycle to give long-term stability to the battery.

a (200), (220) and (400) reflections of synchrotron operando XRPD pattern. b 2D contour plot for operando

XRPD for the first 3 cycles. Structural and Rietveld refinements for NMF cathode following 1st cycle in (c) Alkaline electrolyte and (d) Alkaline electrolyte with Ni/C coating. Comparison of XRPD pattern following 1st cycle and 3rd cycle in (e) alkaline electrolyte and (f) alkaline electrolyte with Ni/C coating.

To assess the possible universality of the new electrode modification method in alkaline batteries, the Co/C nanoparticle was used to build the cathode coating. Similar with Ni nanoparticles, Co can be oxidized to Co(OH)2 in alkaline media and, it exhibits a reversible redox pair of Co(OH)2/CoOOH, together with the ability to in-situ substitute the Mn atom. As a result, good stability of the battery with Co/C coating is achieved (Supplementary Fig. 25). This finding provides evidence for the universality of creating H3O+-rich cathode surfaces and in-situ optimizing the NMF structure by building metal nanoparticle coating to boost the performance of Mn-based PBA cathode in an alkaline environment.

A neutral electrolyte with a concentration of 17 m (mol kg-1) was prepared by dissolving NaClO4 in water. This neutral electrolyte served as the base solution for subsequent preparations. Alkaline electrolytes were obtained by adding 0.1, 0.2, 0.4 and 0.8 mL of 1 M NaOH solution to 30 mL of the neutral electrolyte. For subsequent full cell testing, the optimized alkaline electrolyte prepared with the addition of 0.4 mL of 1 M NaOH was used unless mentioned otherwise.

The coating was prepared as follows: 0.1 g Nafion-Na was dissolved in mixed solution of 0.45 g N, N-Dimethylformamide and 0.45 g isopropanol at 60 ?C. 0.025 g Ni/C and magnetically stirred for 0.5 h, and ultra-sounded for 0.5 h. Procedures were replicated three times to give an even mixture. 10 uL cm-2 solution was sprayed on the surface of the cathode discs. Following removal of the solvent at room temperature (RT) (ca. 25 ?C) in N2-filled glove box under vacuum over 24 h, the electrode discs were coated homogenously.

For DFT computation of the Ni substitution energy, to account for strong correlation effects in the 3d orbitals of Mn, Ni and Fe, a Hubbard U correction was included with the values of 4.0, 5.5 and 4.0 eV, respectively. The kinetic energy cut-off for plane wave expansion was set at 800 eV in all computations.

The initial NMF structure was constructed using 2 x 2 x 2 supercells with Fe and Mn atoms alternatively occupying metal sites. To simulate the dissolution of Mn atom, one Mn atom from the initial NMF structure was intentionally removed before geometry optimization. To simulate the doping with Ni atom, a Mn atom was substituted by a Ni atom in the initial NMF structure, followed by geometry optimization. For geometry optimization, the Brillouin zone was sampled with a (3 x 3 x 3) grid of k-points mesh with a Gamma-centered Monkhorst-Pack scheme. The structure was relaxed until energy and force converged below 10-6 eV and 0.05 eV/?, respectively.