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Epoxy Resin-Reinforced F-Assisted $Na_3Zr_2Si_2PO_{12}$ Solid Electrolyte for Solid-State Sodium Metal Batteries

Yao Fu, Dangling Liu, Yongjiang Sun, Genfu Zhao and Hong Guo *

International Joint Research Center for Advanced Energy Materials of Yunnan Province, Yunnan Key Laboratory of Carbon Neutrality and Green Low-Carbon Technologies, School of Materials and Energy, Yunnan University, Kunming 650091, China
* Correspondence: guohong@ynu.edu.cn

Abstract: Solid sodium ion batteries (SIBs) show a significant amount of potential for development as energy storage systems; therefore, there is an urgent need to explore an efficient solid electrolyte for SIBs. $Na_3Zr_2Si_2PO_{12}$ (NZSP) is regarded as one of the most potential solid-state electrolytes (SSE) for SIBs, with good thermal stability and mechanical properties. However, NZSP has low room temperature ionic conductivity and large interfacial impedance. F^- doped NZSP has a larger grain size and density, which is beneficial for acquiring higher ionic conductivity, and the composite system prepared with epoxy can further improve density and inhibit Na dendrite growth. The composite system exhibits an outstanding Na^+ conductivity of 0.67 mS cm^{-1} at room temperature and an ionic mobility number of 0.79. It also has a wider electrochemical stability window and cycling stability.

Keywords: sodium ion battery; epoxy-NZSPF$_{0.7}$; composite solid electrolyte; solid-state electrolyte

1. Introduction

Energy storage systems (ESSs) are gradually becoming essential and important in people's daily lives, as these can provide us with convenience in many aspects [1–4]. As a hopeful substitute for lithium-ion batteries (LIBs), SIBs have caught the attention of a number of researchers recently because of their rich sodium resources, low prices and excellent sustainability. However, because sodium is more reactive than lithium, it is more likely to form dendrites in the conventional liquid electrolyte battery system, posing serious efficiency problems and safety hazards. Hence, research on solid or quasi-solid sodium ion batteries is of great importance for improving battery efficiency and safety [5–16].

To date, different types of sodium materials, such as Na-β''-Al_2O_3, sulfides, polymers, and Na superionic conductor (NASICON) have been reported for use as sodium ion solid-state electrolytes. As a solid electrolyte, Na-β''-Al_2O_3 is now successfully used in Na-S batteries; however, it is sensitive to moisture [17] and has a high preparation temperature, which poses some limitations to its production applications. Most sulfide-based solid electrolytes are limited in their application due to their instability in air [18] and narrow electrochemical stability window [19], despite their high ionic conductivity and good ductility. The polymer solid electrolyte is flexible, and the contact between it and the electrode is flexible, which makes it malleable and easy to process and shape. However, the ionic conductivity and ion transfer number dose not meet the imposed requirements when using at room temperature. To achieve ionic conductivity for battery applications, a temperature of 60 °C or higher is required. This temperature approaches the melting point of anode, Na (97 °C), which can cause safety problems. By comparing with the sodium-ion solid electrolyte above, a significant amount of attention has been devoted to NASICON ($Na_{1+x}Zr_2Si_xP_{3-x}O_{12}$) because of its high ionic conductivity, wide window of electrochemical stability and stability in air. Hong [20] and Goodenough [21] were the first to study $Na_{1+x}Zr_2Si_xP_{3-x}O_{12}$. The highest ionic conductivity was 10^{-4} S cm^{-1} (x = 2) at room temperature [20,22]. It has also been further improved by doping modifications

of NASICON. The rare earth element La-doped $Na_3Zr_2Si_2PO_{12}$ has an ionic conductivity comparable with that of ionic liquid electrolytes (10^{-1} S cm^{-1}) [23].

In recent years, several studies have reported anion-assisted ways to increase the room temperature's ionic conductivity [23–25]. Lu et al. modified the $Li_{6.25}Ga_{0.25}La_3Zr2O_{12}$ crystals via F$^-$ assisted synthesis to generate smoother and quicker diffusion channels for Li$^+$, thus improving the ionic conductivity [23]. Li et al. reported that anion-substituted $Li_{3x}La_{2/3-x}TiO_3$ has a higher ionic conductivity [24]. Goodenough reports that the substitution of F for OH, which allows $Li_2(OH)_{0.9}F_{0.1}Cl$ to have an improved Li$^+$ diffusion path [26]. Although anion-assisted NZSPs have high ionic conductivity, they still do not provide effective inhibition of dendrite growth, resulting in uneven Na plating or streaking, leading to poor cell performance. This is because voids still exist on the surface of the SSE, leading to the uneven deposition of Na on the electrode surface and promoting the growth of dendrites. Here, we investigated F$^-$-assisted NZSP solid electrolyte materials. It was found that the grain size and densities of NZSP increased with the introduction of F. However, it is still not enough to inhibit the growth of Na dendrites. Then, we prepared a composite system by depositing epoxy into the pores of NZSPF via vacuum adsorption. The composite system greatly modified the cycling performance of the cell with almost no decrease in ionic conductivity and no increase in impedance. The ionic conductivity of epoxy-NZSPF$_{0.7}$ is 0.67 mS cm^{-1} (NZSPF$_{0.7}$ is 0.95 mS cm^{-1}). In addition, epoxy-NZSPF$_{0.7}$ has a wider electrochemical stability window. Finally, we also assembled a Na | epoxy-NZSPF$_{0.7}$ | $Na_3V_2(PO_4)_3$ quasi-solid SIB, with good cycling performance at 40 °C.

2. Materials and Methods

2.1. Synthesis of NZSPF$_x$ Solid Electrolyte

Traditional solid-phase reactions were adopted to synthesize F$^-$-assisted NZSPF$_x$ materials. Na_2CO_3, ZrO_2, SiO_2, and $NH_4H_2PO_4$ were weighed according to certain stoichiometric ratios and then x mol of NaF (x = 0, 0.1, 0.3, 0.5, 0.7, and 1.0) was added separately. The final results were labeled NZSPF$_x$. Balls, materials and ethanol were added to the ball mill tank in the ratio of 32:5:8. The milling was then carried out with a planetary ball mill at 400 rpm for 10 h. The 12 h drying of precursors was carried out under vacuum at 80 °C; then, the mixtures were transferred to a muffle furnace for 12 h preheating at 900 °C, followed by 3 h sintering at 1050 °C. The sintered powders were further ball-milled at 200 rpm for 10 h to obtain a uniform powder size. During the ball-milling process, a 5% mass fraction of PVA solution was added to act as a binder. The pressing of powers into pellets (diameter 16 mm, thickness 1 mm) was carried out at 120 MPa. Finally, the 4 h sintering of pellets was carried out at 800 °C to remove the PVA solution. After cooling to room temperature, the 24 h sintering of pellets was performed at 1100 °C to obtain NZSPF$_x$ solid electrolyte.

2.2. Synthesis of Epoxy-NZSPF$_{0.7}$ Solid Electrolyte

The DGEBA (diglycidyl ether of bisphenol-A) and PACM (4,4'-diaminodicyclohexyl-methane) were dissolved in THF in stoichiometric ratios to obtain 1 mol L^{-1} solutions, respectively. Then, the two solutions were mixed in a volume ratio of 2:1 (DGEBA:PACM) and stirred for 1 h. The sintered NZSPF ceramic sheets in 2.1 were completely immersed in the mixed solution, kept in a vacuum environment for 20 min and cycled three times. The sintering of pellets was carried out at 150 °C for 24 h to obtain an epoxy-NZSPF$_x$ solid electrolyte.

2.3. Characterization and Measurements

A Bruker D8 Advance X-ray diffractometer configured with Cu Kα radiation in the 2θ scope of 10–60° was adopted to collect the X-ray diffraction (XRD) patterns, and the collection of data was carried out at 5° min^{-1}. The chemical component in an Escalab 250Xi instrument from Thermo Scientific configured with an Al Kα micro-focused X-ray

source was demonstrated via X-ray photoelectron spectroscopy (XPS) tests. The collection of Fourier transform infrared (FTIR) spectra were carried out on a Nicoletis 10 spectrometer from Thermo Scientific. The spectrum was from 4000 cm^{-1} to 400 cm^{-1}. The microstructures of SSE were studied using scanning electron microscopy (SEM) (QUNATA-FEG). The measurement of thermostability was carried out via thermogravimetric analysis (TGA) on a Netzch STA449F3 analyzer under N_2 conditions at a heating rate of 10 °C/min from 25 to 700 °C.

A CHI660E (ChenHua) electrochemical workstation with an AC amplitude of 10 Mv and a frequency scope from 10 Hz to 10^6 Hz was adopted to make electrochemical impedance spectroscopy (EIS) tests at room temperature. A combination of direct-current (DC) polarization and alternating-current (AC) impedance was used for the ion transference number test. A CR2032 cell case was chosen to assemble a Na/NZSPF$_x$/Na symmetric battery. AN AC impedance test is performed on the battery before the DC polarization test; then, a small bias voltage (5 mV) is added to the battery for the DC polarization test, and after the planned current of the battery is stabilized, the battery is then tested for AC impedance again. The electrochemical stability windows of NZSPF$_{0.7}$ and epoxy-NZSPF$_{0.7}$ solid electrolytes were evaluated with cyclic voltammetry (CV) and linear sweep voltammetry (LSV). The tests were performed on the Au | NZSPF$_{0.7}$ | Na and Au | epoxy-NZSPF$_{0.7}$ | Na cells and the scanning rate was 0.2 mV s^{-1} at room temperature. The electrochemical stability test was carried out to test the interfacial stability and Na stripping–plating behavior. The testing of assembled Na | NZSPF$_x$ | Na and Na | epoxy-NZSPF$_{0.7}$ | Na symmetric cells was carried out on a Blue Power Test System CT2001A with a constant current density of 0.1 mA cm^{-1} at 40 °C.

The assembly of the full cell was carried out in an argon atmosphere glove box using Na metal as the anode, NZSPF$_{0.7}$ and epoxy-NZSPF$_{0.7}$ as the solid electrolyte, and $Na_3V_2(PO_4)_3$ as the cathode using a 2032 cell case. For the preparation of the cathode electrode sheet, $Na_3V_2(PO_4)_3$, the mixture of acetylene black carbon and PVDF was carried out in NMP with a mass ratio of 7:2:1. Using Al foil as a fluid collector, the coasting of the slurry was carried out on the surface of Al foil, followed by 12 h OF drying under vacuum at 80 °C to eliminate the NMP solvent. The Na metal, solid electrolyte and cathode pole piece were assembled in the order of Na | epoxy-NZSPF0.7 | $Na_3V_2(PO_4)_3$. The addition of 10 μL liquid electrolyte (ethylene carbonate (EC) as the solvent and $NaClO_4$ as the salt) was conducted between the solid electrolyte and the pole pieces to wet the contact surface. The constant current charge/discharge test of the batteries was performed on the LAND CT2001A test system at 40 °C to evaluate the long-cycle performance.

3. Results and Discussion

To investigate the influence of adding F$^-$, we used XRD to measure the lattice structure of samples with different fluorine contents. According to Figure S1, the main diffraction peaks of NZSPF$_x$ (x = 0, 0.1, 0.3, 0.5, 0.7, and 1.0) are consistent with the monoclinic NZSP structure [27]. This phenomenon proves that the introduction of F$^-$ did not destroy the crystal structure. However, with the increase in NaF content, a ZrO_2 secondary phase was detected. This may be due to the reaction of Si^{4+}/P^{5+} with F during the process of high-temperature sintering, as it leads to a reduction in Si^{4+}/P^{5+} content and, finally, the deposition of the ZrO_2 secondary phase. In addition, no fluorine-related phases were observed in the XRD patterns of all samples, which could be due to F occupying positions in the lattice dot matrix or high-temperature volatilization.
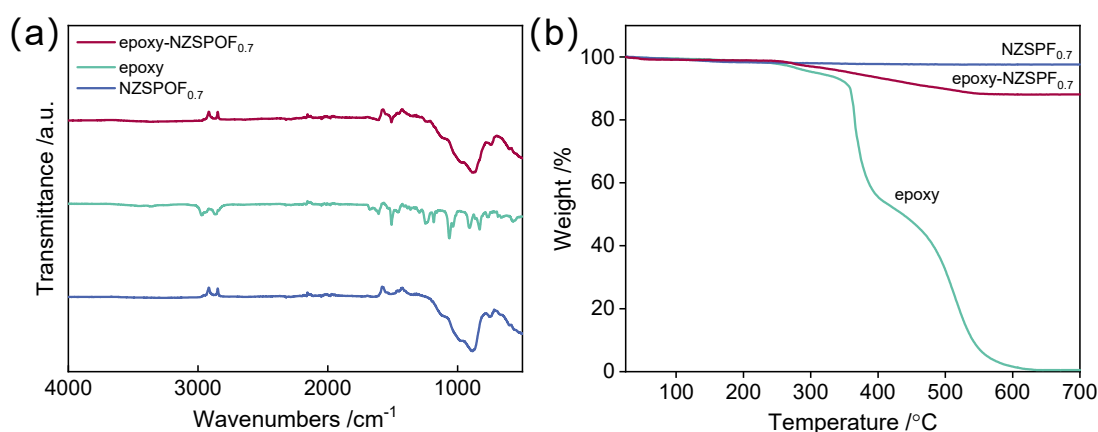
These featured peaks of P-O and Si-O groups (Figure S2) were analyzed with FTIR spectroscopy. All samples displayed the same stretching or bending vibration pattern. The peaks at 598 and 1128 cm^{-1} were ascribed to P-O stretching in tetrahedral PO_4^{3-} units [25,27], while the peaks at 501 and 863 cm^{-1} were ascribed to the Si-O stretching in tetrahedral SiO_4^{4-} units [25,28]. These outcomes definitively prove that SiO_4^{4-} and PO_4^{3-} units are in the NZSPF$_x$ crystal structure. These peaks are shifted to varying degrees,

presumably due to F occupying the position of the O element in the $NZSPF_x$ lattice, causing the conformity of positive and negative centers in the lattice [26].

To further investigate the elemental composition of $NZSPF_x$, XPS was used to characterize NZSP, $NZSPF_{0.7}$. Figure S3a shows the full XPS spectra of NZSP and $NZSPF_{0.7}$. Different peak areas corresponding to Na, Zr, Si, P, O are obviously found, which proves that the major components of the ceramics is $NZSPF_{0.7}$. High-resolution XPS (Figure S3b) is adopted to detect the F 1s peak, which proves the successful doping of the F element. The binding energy at 684 eV corresponds to the $F-Si/P$ bond [29,30], showing that the F^- takes up the O^{2-} place in the $NZSPF_x$ lattice. The intensities of the P 2p and Si 2p peaks reduce slightly under the help of F^- (Figure S3c,d), indicating a slight decrease in the concentration of P^{5+} and Si^{4+} because of the reaction of P^{5+}/Si^{4+} with F and the volatilization of the results at high temperatures. In addition, a slight shift in P 2p and Si 2p to a higher binding energy was observed in $NZSPF_{0.7}$ by comparing with the undoped sample, and the presence of $P/Si-F$ bonds [31] can explain this shift, which also proves that F^- takes up part of the O^{2-} sites in the $NZSPF_{0.7}$ lattice.

Although the doping of NaF can reduce the resistance of NZSP (Figure S5) and improve the ionic conductivity (Figure S6), the cycling stability is deemed unsatisfactory (Figure S7). This is because the $NZSPF_{0.7}$ grain boundaries are not dense, which also leads to the ability of growing and forming Na dendrites in the crevices (Figure S9). Therefore, it is necessary to modify $NZSPF_{0.7}$ to strengthen the interface between the SSE and the electrode, and to stop the development of Na dendrites.

Figure 1a shows the FTIR spectra of the epoxy, $NZSPF_{0.7}$, and epoxy-$NZSPF_{0.7}$ composite system. The FTIR spectra of epoxy-$NZSPF_{0.7}$ do not show new peaks, only a simple superposition of the two monomers. This indicates that no new substances are produced in the composite system and no reaction occurs between the two monomers. Figure 1b shows the TGA curves of epoxy, $NZSPF_{0.7}$, and epoxy-$NZSPF_{0.7}$. It can be seen that in the tested temperature range, $NZSPF_{0.7}$ does not undergo mass loss and is thermally stable, while the mass loss of epoxy starts to occur at 300 °C and almost completely decomposes after reaching 600 °C. This coincides with the weight loss range of the composite system, and it can be determined that the mass loss of the composite system is caused by epoxy. This indicates that epoxy was successfully filled into the voids of $NZSPF_{0.7}$, further improving the densities of the ceramics (Figure S10).

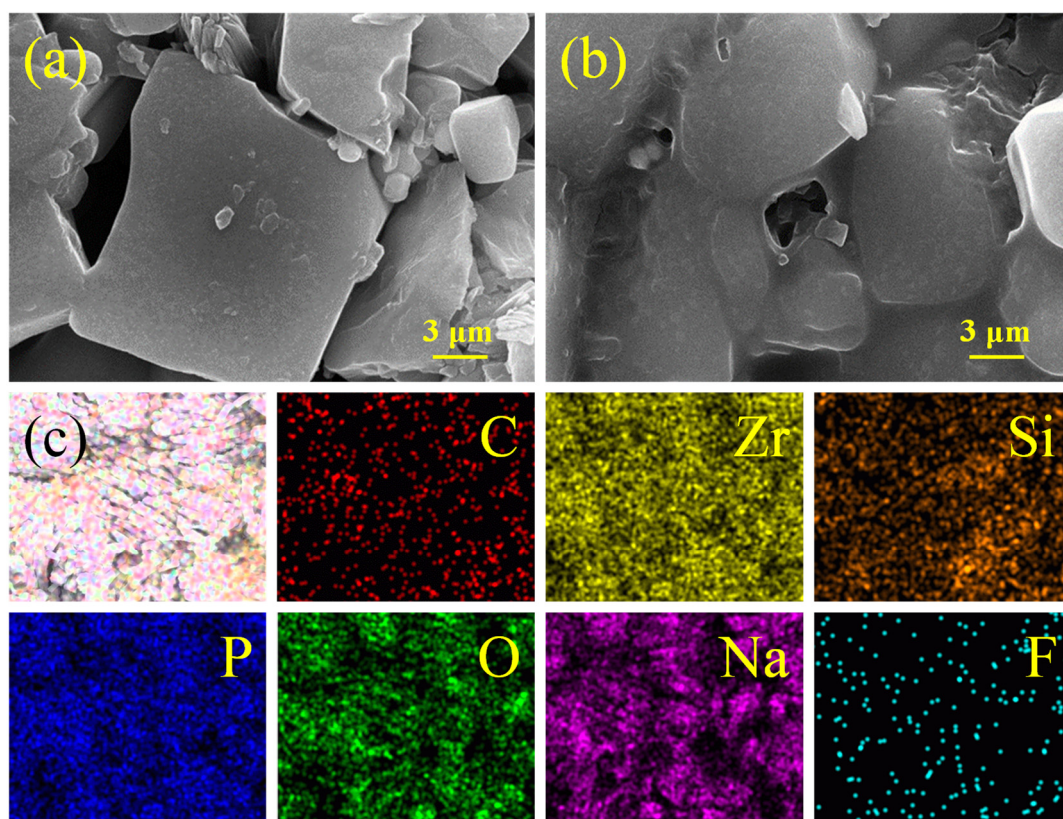**Figure 1.** (**a**) FTIR spectra and (**b**) TGA analysis for $NZSPF_{0.7}$, epoxy and epoxy$-NZSPF_{0.7}$.

To observe the surface difference between $NZSPF_{0.7}$ and epoxy-$NZSPF_{0.7}$, and the distribution of epoxy resin in $NZSPF_{0.7}$, SEM and EDS tests were performed on both $NZSPF_{0.7}$ (Figure S4) and epoxy-$NZSPF_{0.7}$ (Figure 2). Figure S4 shows the SEM image of $NZSPF_{0.7}$. The SEM images show that with the growth in NaF content, the size of $NZSPF_x$ grains gradually grows larger. However, F promotes grain growth, reduces grain boundary concentration and increases ceramic density to a certain extent, which can efficiently decrease the grain boundary concentration, increase the ionic conductivity and reduce
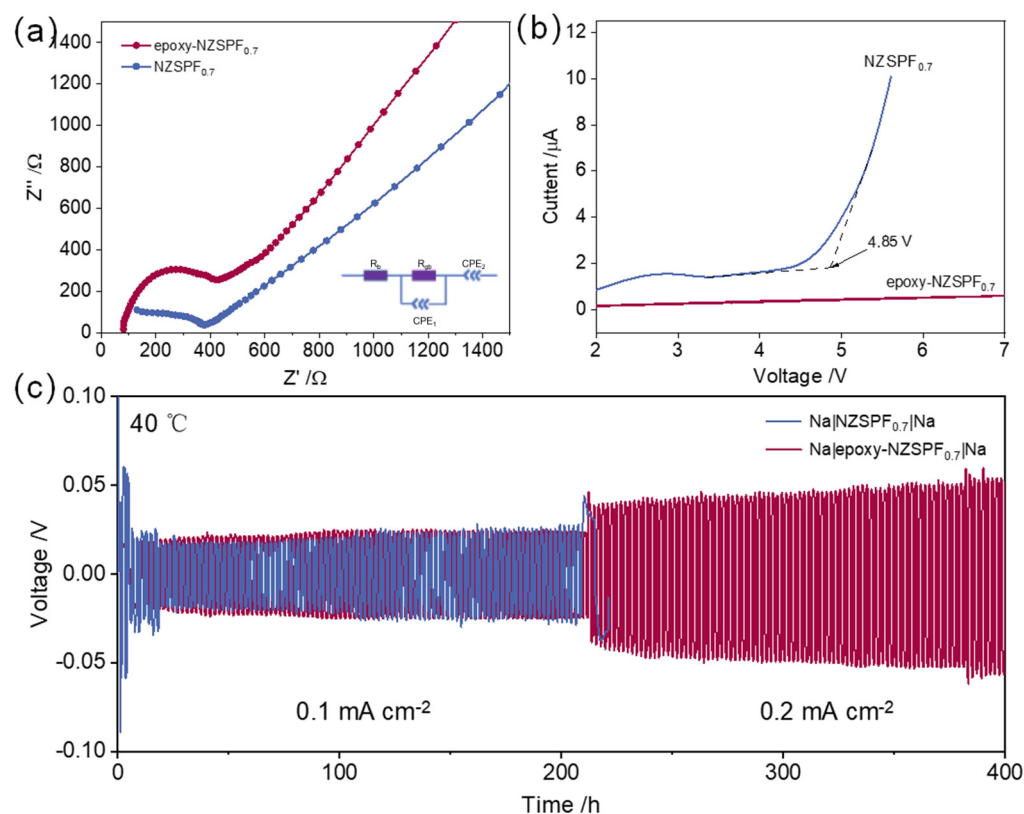
the grain boundary resistance [32,33]. There are still a large number of interstices in the middle of the ceramic after high-temperature sintering, and the existence of these interstices leads to uneven deposition of dendrites here and reduces the electrochemical performance. Figure 2 shows the surface of epoxy-NZSPF$_{0.7}$ composite solid electrolyte after polishing; compared with NZSPF$_{0.7}$, the gap in the composite system is significantly reduced, which is a good verification of the increased densities. The denseness of epoxy-NZSPF$_{0.7}$ is 97.6% and NZSPF$_{0.7}$ is 93.4%, while that of un-assisted NZSP is only 79.8%. This shows that the filling of epoxy can raise the densities of the composite system because the prior condition for a solid electrolyte to have a high ionic conductivity is a high density [34,35]. Additionally, Figure 2c displays the EDS discussion on the epoxy-NZSPF$_{0.7}$ ceramic sample. Because epoxy contains a large number of C elements, by observing the distribution of C elements, we can find that epoxy is evenly distributed in the composite system. The maps also confirm that O, P, Si, F, Na and Zr factors generally show an even homogeneous distribution in the composite system, and the content of the F element is essentially the same as that of NASPF$_{0.7}$.

Figure 2. SEM image of (**a**) NZSPF$_{0.7}$ and (**b**) epoxy-NZSPF$_{0.7}$; (**c**) EDS mapping of epoxy-NZSPF$_{0.7}$.

The electrical conductivities of the solid electrolyte were investigated with EIS. Figure 3a displays the impedance spectra of NZSPF$_{0.7}$ and epoxy-NZSPF$_{0.7}$. According to the Nyquist curve, the resistance of NZSPF$_{0.7}$ and epoxy-NZSPF$_{0.7}$ are calculated as 370 Ω and 420 Ω. The overall conductivity measures from the EIS are 0.95 and 0.67 mS cm^{-1}. The impedance of epoxy-NZSPF$_{0.7}$ becomes larger and the ionic conductivity decreases are due to the fact that the epoxy impedance of the pure phase is extremely large; therefore, filling the gap to NZSPF$_{0.7}$ results in a growth in the impedance and a decline in the ionic conductivity of epoxy-NZSPF$_{0.7}$.

Figure 3. (**a**) EIS measurements at room temperature; (**b**) LSV of the Au | NZSPF$_{0.7}$ | Na and Au | epoxy−NZSPF$_{0.7}$ | Na cells at room temperature; (**c**) variable current cycling of Na | NZSPF$_{0.7}$ | Na and Na | epoxy−NZSPF$_{0.7}$ | Na symmetric cells (the current density is 0.1 mA cm^{-2} and becomes 0.2 mA cm^{-2} after 200 h).

To verify the conduction behavior of sodium ions, the sodium ion transfer number (t_{Na}^+) was determined. The t_{Na}^+ (40 °C) of NZSPF$_{0.7}$ was 0.84, while that of epoxy-NZSPF$_{0.7}$ was 0.79. This proves that the filling of epoxy does not significantly affect the t_{Na}^+ and Na$^+$ is still transported by NZSPF$_{0.7}$ as a transport channel rather than epoxy.

In addition to having a high ionic conductivity, a wide-range electrochemical stability window is also a basic necessity for SSE in actual use. The electrochemical stability windows of NZSPF$_{0.7}$ and epoxy-NZSPF$_{0.7}$ is assessed via LSV with Au | NZSPF$_{0.7}$ | Na and Au | epoxy-NZSPF$_{0.7}$ | Na batteries with a scan rate of 0.2 mV s^{-1} at 30 °C (Figure 3b). The electrochemical stability window of the NZSPF$_{0.7}$ is 4.85 V, while the current intensity of epoxy-NZSPF$_{0.7}$ remains constant in the range of 2–7 V. This indicates that the electrochemical steadiness window of theepoxy-NZSPF$_{0.7}$ up to 7 V. It is expected that epoxy-NZSPF$_{0.7}$ can be used as a high-voltage sodium battery. The electrochemical properties of NZSPF$_{0.7}$, NZSPF$_{0.7}$ and epoxy-NZSPF$_{0.7}$ are listed in Table 1.
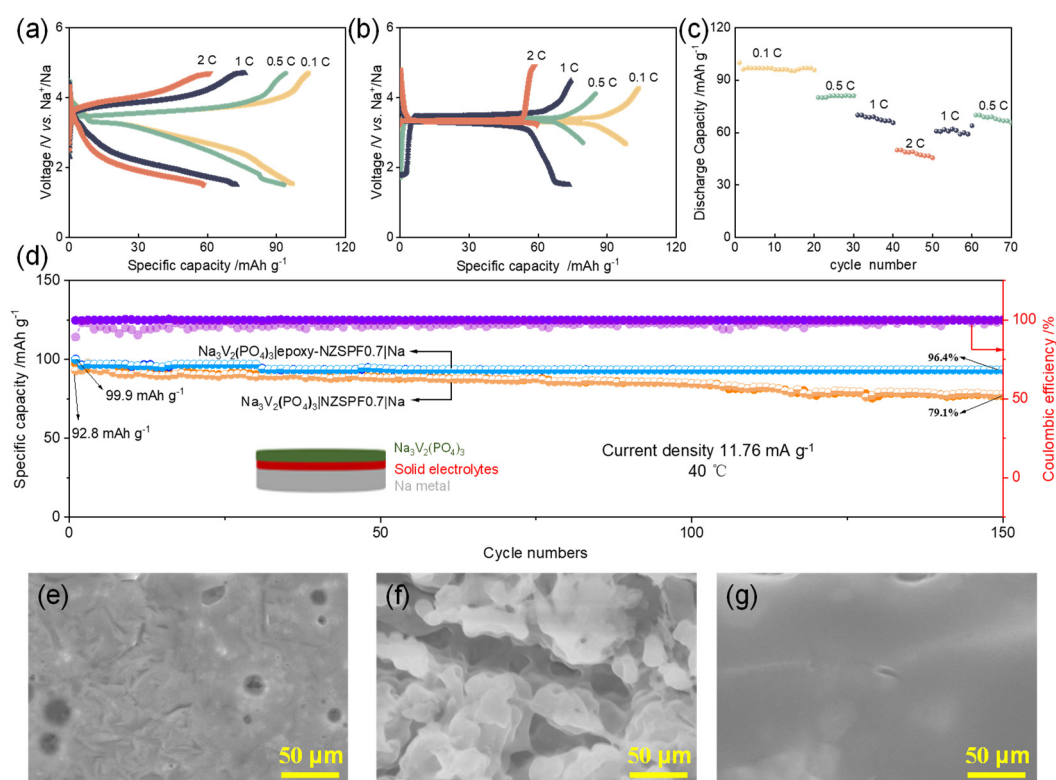
Table 1. Electrochemical properties of NZSPF$_0$, NZSPF$_{0.7}$ and epoxy-NZSPF$_{0.7.}$

Sample	Impedance at Room Temperature	Ionic Conductivity	Ion Transfer Number	Electrochemical Stabilization Window
NZSPF$_0$	1527	0.45	0.68	-
NZSPF$_{0.7}$	370 Ω	0.95 mS cm^{-1}	0.84	4.85 V
epoxy-NZSPF$_{0.7}$	420 Ω	0.67 mS cm^{-1}	0.79	7 V

Figure 3c shows that epoxy-NZSPF$_{0.7}$ can be cycled steadily for above 200 h (100 cycles) at a current density of 0.1 mA cm^{-2} with no fluctuation in potential, and remains stable for nearly 200 h when the current density grows to 0.2 mA cm^{-2}. This indicates that

epoxy-NZSPF$_{0.7}$ can promote the even deposition of Na$^+$ and stop the Na dendrites from developing. In contrast, NZSPF$_{0.7}$ could only keep steady at 0.1 mA cm^{-2} and immediately short-circuited at 0.2 mA cm^{-2}. It can be noted that NZSPF$_{0.7}$ needs 5–10 h to adapt to the electrode with significant potential fluctuations, while epoxy-NZSPF$_{0.7}$ hardly needs this process. Compared to this, epoxy-NZSPF$_{0.7}$ requires a shorter activation time and does not show significant short-circuiting after increasing the current, indicating that it has higher electrode stability.

Finally, Na | NZSPF$_{0.7}$ | Na$_3$V$_2$(PO$_4$)$_3$ and Na | epoxy-NZSPF$_{0.7}$ | Na$_3$V$_2$(PO$_4$)$_3$ solid SIBs were assembled. Figure 4 shows the performances of batteries. The polarization of NZSPF$_{0.7}$ is bigger with the gradual current rate growth (Figure 4a), while the charge/discharge curve of epoxy-NZSPF$_{0.7}$ is smooth and the polarization is basically unchanged (Figure 4b). The discharge specific capacities of epoxy-NZSPF$_{0.7}$ are 96.9, 80.1, 68.9 and 48.9 mAh g^{-1} at 0.1C, 0.5C, 1C and 2C. In the case of the reversion of the current density to 1C and 0.5C, the given capacities are 60.8 and 70.5 mAh g^{-1} (Figure 4c). This shows that when the current returns to the same rates, it has excellent reversibility and steadiness, although the reversible capacity is slightly reduced. Figure 4d shows the cycling performance of cells assembled with two different electrolytes at a current rate of 0.1C at 40 °C. The discharge-specific capacity remains 93.7 mAh g^{-1} after 150 cycles, which is 95.8% capacity retention of initial capacity (97.8 mAh g^{-1}). The mean coulombic efficiency is close to 99%, which is only 79.1% for Na$_3$V$_2$(PO$_4$)$_3$ | NZSPF$_{0.7}$ | Na battery.

Figure 4. (**a**) Charging–discharging profiles of Na$_3$V$_2$(PO$_4$)$_3$ | NZSPF$_{0.7}$ | Na at different ates; (**b**,**c**) charging–discharging profiles of Na$_3$V$_2$(PO$_4$)$_3$ | epoxy-NZSPF$_{0.7}$ | Na at different rates; (**d**) cycling performance of the Na$_3$V$_2$(PO$_4$)$_3$ | epoxy−NZSPF$_{0.7}$ | Na battery at a current density of 11.76 mA g^{-1}; (**e**) SEM image of sodium metal surface before cycling; SEM image of sodium metal surface of (**f**) Na$_3$V$_2$(PO$_4$)$_3$ | NZSPF$_{0.7}$ | Na battery and (**g**) Na$_3$V$_2$(PO$_4$)$_3$ | epoxy−NZSPF$_{0.7}$ | Na battery after cycling.

Figure 4e–g shows the SEM images of the Na electrode surface. Obviously, after 150 cycles, Na dendrites were formed at the boundaries and voids of the NZSPF0.7 solid electrolyte (Figure 4f), resulting in inhomogeneous Na plating/striping with a corre-

sponding aggravation of the cell behavior [30]. However, the Na electrode surface of $Na_3V_2(PO_4)_3$ | epoxy-NZSPF$_{0.7}$ | Na battery was clean and smooth (Figure 4g), and almost no Na deposition was found, indicating that the epoxy-filled dielectric material can effectively prevent the formation of dendrites and enhance the battery cycling behavior.

4. Conclusions and Outlook

In conclusion, the grain boundary concentration of NZSP was effectively reduced by introducing NaF into the NZSP solid electrolyte and a denser NZSPF$_{0.7}$ with improved ionic conductivity was obtained (0.98 mS cm^{-1}); however, NZSPF$_{0.7}$ did not stop the development of sodium dendrites well and had poor interfacial properties. For this reason, a composite solid electrolyte epoxy-NZSPF$_{0.7}$ was prepared via simply vacuum adsorption and by filling the interstitial space of NZSPF$_{0.7}$ with epoxy and curing it. Epoxy-NZSPF$_{0.7}$ combines the great ionic conductivity of inorganic electrolytes as well as the good interfacial contact from organic polymer electrolytes, with high ionic conductivity (0.67 mS cm^{-1}) as well as excellent interfacial performance and cycling behavior (the initial capacity of cells is 97.9 mAh g^{-1} with a 96.4% retention rate after 150 turns). According to the above-presented results, this study may offer a novel idea for research concerning solid electrolytes for sodium-ion batteries.

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