



# Facile preparation and characterization of poly(vinyl alcohol)-NaCl-glycerol supramolecular hydrogel electrolyte

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## ABSTRACT

The facile preparation of hydrogel electrolyte with high strength, healing ability and thermoplasticity is important and still remains a great challenge. In this paper, a novel way for the preparation of poly(vinyl alcohol) (PVA) supramolecular hydrogel electrolyte was presented. The PVA hydrogel electrolyte was prepared via the hydrogen bonding and salting-out effect. The high performance PVA-NaCl-glycerol hydrogel electrolyte could be obtained by the doping of glycerol and NaCl into PVA aqueous solution and storing at room temperature for 2.0 h. The crystallinity, thermal and mechanical properties of PVA-NaCl-glycerol hydrogel electrolyte were characterized by Fourier transformed infrared spectroscopy, X-ray diffraction measurement and tensile testing, respectively. The hydrogel forming mechanism was discussed. The PVA-NaCl-glycerol hydrogels performed the high toughness with the tensile strength and elongation at break of  $0.57 (\pm 0.02)$  MPa and  $575 (\pm 29)\%$ , respectively. The conductivity of PVA-NaCl-glycerol was as high as  $92.5 (\pm 0.18)$  mS/cm. The PVA-NaCl-glycerol hydrogel electrolyte behaved the excellent heat healing ability and thermoplasticity. These properties of PVA-NaCl-glycerol are related to the role of glycerol and NaCl. This paper provides a new preparation method and structure design idea for the high performance hydrogel electrolyte.

## 1. Introduction

Due to the small size, low weight, ease of handling, and excellent reliability, flexible device such as flexible semi-solid-state electric double-layer supercapacitors and flexible lithium ion battery has been a mainstream direction in modern electronics and relevant multi-disciplinary fields [1–6]. One of the critical factors for attaining highly flexible devices is the electrolyte. The rapid development of flexible energy devices has dramatically increased the requirement for hydrogel polymer electrolyte with mechanical robustness, healable property and high excellent transport efficiency of ion or solute [7–11]. However, most of the hydrogels are of poor mechanical properties and cannot be remolded. It still is a challenge to design and synthesize hydrogel electrolyte with these superior properties. To overcome these problems, considerable attention has been focused on improving the abilities of hydrogel electrolyte for application in energy devices, such as aqueous solar cells and aqueous batteries [12–17].

Hydrogel polymer electrolyte can be made from natural and synthetic polymers. The polymeric network of hydrogels is always formed through covalent interactions between different polymer chains by cross-linkers or through noncovalent interactions such as hydrogen bonding [18], ionic interactions [19–21], host–guest interactions or a

combination of these interactions [22]. Poly(vinyl alcohol) (PVA) is an important water soluble synthetic polymer as a green material. Due to its biodegradability and high robustness [23], PVA hydrogel is a widely used GPE substrate. PVA hydrogel can be prepared through either physical crosslinking or chemical crosslinking processes. Freezing/thawing method is the typical example of physical crosslinking [24–27]. The mechanical properties of PVA hydrogels usually can be improved by increasing the freezing/thawing cycles to form micro-crystallines in the hydrogel as the physical crosslinking points. For chemical crosslinking the crosslinking points are formed between PVA chains by adding chemical crosslinker to improve the mechanical properties of PVA hydrogels.

Currently, the PVA conducting hydrogel usually consists of PVA chains and inorganic salt, acid or alkali dissolved in water. However, these hydrogels usually are brittle and exhibit low mechanical strength due to the damage effect of inorganic salt to the hydrogen bonding between PVA chains and water molecules. Moreover, the metal electrodes would be ruined by the acid or alkali in PVA hydrogel electrolyte. Thus, the PVA hydrogel with neutral salts has attracted the attention of many researchers [28]. However, the salt content in the hydrogel electrolyte is usually low (generally lower than 20 wt%) due to the limited compatibility between the polymer matrix and salt.

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**Table 1**

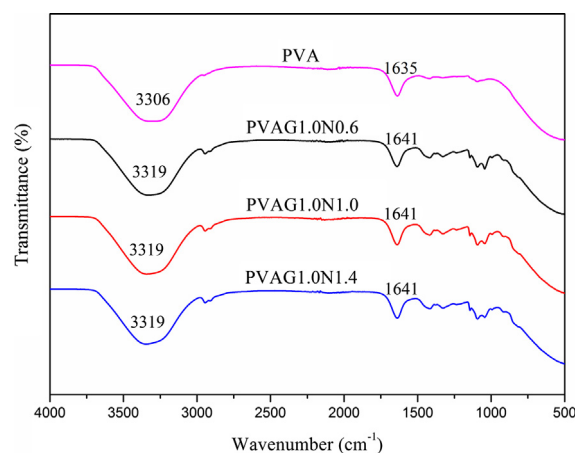
The experimental ingredients and nomenclatures of PVA-NaCl-glycerol hydrogel electrolyte.

Samples	PVA (g)	Glycerol (g)	NaCl (g)	Distilled water (g)
PVA	1.0	/	/	7.0
PVAG1.0N0.6	1.0	1.0	0.6	7.0
PVAG1.0N0.8	1.0	1.0	0.8	7.0
PVAG1.0N1.0	1.0	1.0	1.0	7.0
PVAG1.0N1.2	1.0	1.0	1.2	7.0
PVAG1.0N1.4	1.0	1.0	1.4	7.0

Therefore the ionic conductivities of hydrogel electrolyte are generally low (0.1–10 mS/cm). The low conductivity and poor mechanical strength would limit the application of PVA hydrogel.

The previous studies showed that PVA tough hydrogel could be obtained at room temperature through simple physical mixing of PVA and glycerol in water. In this system, water and small glycerol molecules would function as bridge molecules to connect PVA chains into bundles by forming many hydrogen bonds with PVA chains. This work would provide a simple yet physical way to prepare PVA tough hydrogel at room temperature without chemical crosslinker and freezing process. However, the high concentration of glycerol is needed [29].

To our knowledge, no research on the synergistic effect of NaCl and glycerol on the properties of PVA was reported. In this paper, the “salting out” effect and “hydrogen bonding complex” method were used to fabricate the tough hydrogel electrolyte. A novel PVA-NaCl-glycerol flexible hydrogel electrolyte with high conductivity and considerable mechanical properties was prepared by a facile gelation process at room temperature. In addition, PVA-NaCl-glycerol hydrogel electrolyte also possessed excellent healing ability, thermoplasticity and moldability. The presence of these properties in PVA-NaCl-glycerol flexible hydrogel electrolyte can enrich the applications of this material. We stress here that this PVA-NaCl-glycerol flexible hydrogel electrolyte are prepared without the chemical crosslinker and not through the repeated freezing-and-thawing processes. The excellent and easy gelation capability of the



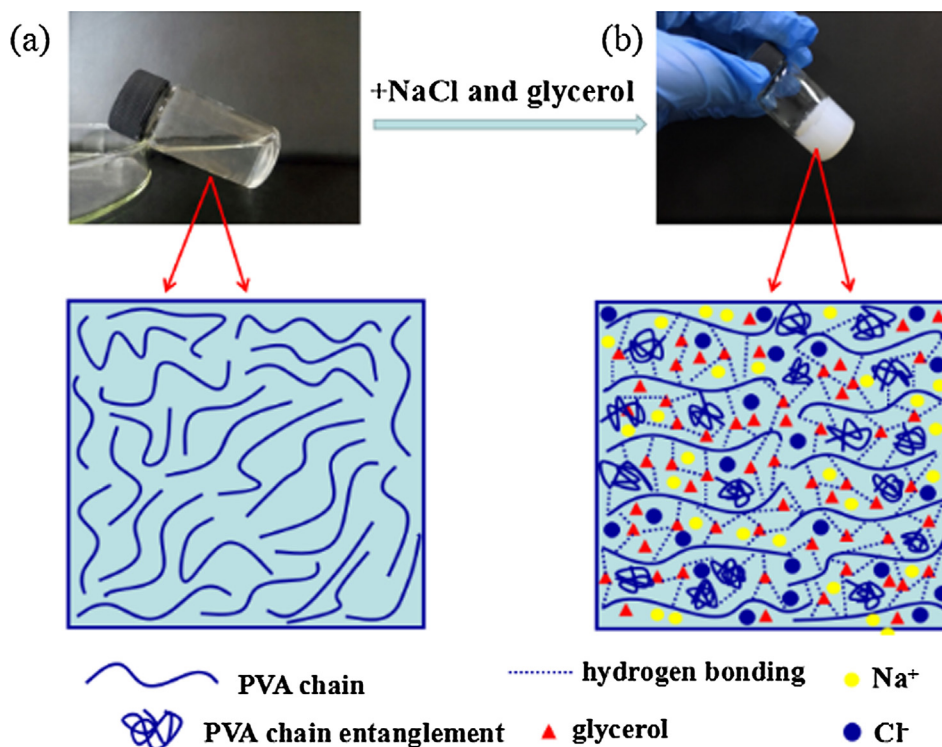
**Fig. 2.** FTIR spectra of various PVA hydrogels: PVA, PVAG1.0N0.6, PVAG1.0N1.0 and PVAG1.0N1.4.

PVA-NaCl-glycerol hydrogel electrolyte at room temperature was achieved by the physical mixing. We hope that this would provide a novel way for the preparation of PVA based hydrogel and enlarge the application of PVA hydrogel.

## 2. Materials and methods

### 2.1. Materials

PVA (1799,  $M_w$ : ~75,000 g/mol, 99% hydrolysis) was purchased from Sichuan Vinylon Factory. NaCl was provided by Sinopharm Chemical Reagent Co. Ltd. Glycerol was purchased from Kelong Chemical Co. (Chengdu, China). Distilled water was used throughout the experiment.



**Fig. 1.** The photos and mechanism of PVA-NaCl-glycerol hydrogel electrolyte forming process: (a) PVA aqueous solution; (b) PVA-NaCl-glycerol hydrogel electrolyte after storing at 25 °C for 2 h.

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