



# Sodium lignosulfonate doped polyvinyl alcohol-based hydrogel conductive composites based on ion-specific effects with continuously tunable mechanical properties and excellent conductive sensing properties

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## ABSTRACT

Hydrogels have great application prospects in the fields of bionics, soft tissue probes, and motion detection, etc. However, the high water content and loose cross-linker structure of hydrogels lead to weak mechanical properties, which limits their application and development. Therefore, in this study, sodium lignosulfonate doped polyvinyl alcohol hydrogels (PVA/LS) were constructed using polyvinyl alcohol as the matrix and sodium lignosulfonate as the filler; the hydrogels were also treated with different ionic solutions to construct ion-responsive hydrogels (PVA/LS/ION); and ion-conducting hydrogels (PVA/LS/Cl) were constructed by the change of the concentration of sodium chloride (NaCl) solution. The hydrophilicity of the polyvinyl alcohol (PVA) molecular chain was changed by different ions, resulting in either water absorption (salting in) or dehydration (salting out), and the hydrogel mechanical properties could be adjusted over a wide range (tensile strength: 77 kPa ~ 3.26 MPa; elongation at break: 290 ~ 870%; modulus: 24 ~ 563 kPa; toughness: 96 kJ/m<sup>3</sup> ~ 14.1 MJ/m<sup>3</sup>). In addition, the hydrogels have good conductivity regulation (1.09 ~ 2.72 S/m) and sensing properties, with GF values up to 1.23, and sensitive and stable strain sensing properties. This work provides useful insights into the application of ion-specific effects to the field of hydrogels.

## 1. Introduction

In recent years, flexible electronics have been widely used in soft robots[1], wearable sensors[2], electronic skins[3], and flexible energy storage devices[4]. Although conventional metal-based electronics show fast response and signal conversion, they usually have poor mechanical flexibility[5]. For biological applications, stretchable electronic materials need to have good electrical conductivity as well as excellent mechanical properties to be suitable for long-term biocompatibility with human tissues such as skin[6]. Hydrogel is a three-dimensional cross-linked polymeric material with a high water content that can be swelled in water and it is in a state between solid and liquid[7]. The polymer network of the hydrogel makes it solid, while the aqueous phase allows the carrier to diffuse rapidly, which indicates that the hydrogel has liquid-like transport properties. Due to this favorable property, hydrogels have good potential for applications in tissue engineering[8], drug release[9], flexible electronics[10,11], wound dressing[12,13], energy storage devices[14,15], adhesives[16] and, soft robots[17].

However, there are still some urgent problems to be solved in the

practical application of hydrogels. First of all, due to the high water content and loose cross-linking structure of hydrogels, their mechanical properties are weak, limiting their potential practical application scenarios. Although current researchers have enhanced the mechanical properties of hydrogels by constructing dual-network hydrogels[18], adding nanofillers[19,20], and mechanical training[21] to extend their range of use. However, compared with anhydrous polymers, their mechanical properties are still unsatisfactory[22]. Secondly, hydrogels are the most desirable materials for tissue engineering applications due to their good biocompatibility[23]. This requires that the modulus of hydrogel is similar to that of biological tissue to promote adhesion to biological tissue[24]. Currently, the modulus of hydrogels has been adjusted by varying the compositions[25], concentrations[26], etc., and these methods have some obvious drawbacks, such as the inability to adjust the modulus in a continuous manner, and a limited range of adjustment. Finally, in some application scenarios, hydrogels need to switch between hard and soft states, such as soft tissue probe applications[27].

Different types of ions exhibit different properties due to differences

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