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Highly stretchable and tough double network hydrogels via molecular stent

Xue Jiao Liu, Xiu Yan Ren *, Shuang Guan, Hai Qiang Li, Zhong Kai Song, Guang Hui Gao *

Engineering Research Center of Synthetic Resin and Special Fiber, Ministry of Education, and Advanced Institute of Materials Science, Changchun University of Technology, Changchun 130012, People's Republic of China

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ABSTRACT

Double network (DN) hydrogels have drawn much attention due to their high mechanical strength and toughness with high-water content. In this investigation, we successfully prepared extremely stretchable, transparent and tough DN hydrogels by using neutral synthetic polymer–poly(vinyl alcohol) (PVA) as the first network, polyacrylamide (PAM) as the second network and sodium alginate (SA) as a molecular stent. It was found that the content of SA significantly influenced the swelling property of PVA semi-interpenetrating polymer network (semi-IPN) hydrogel in acrylamide aqueous media and corresponding mechanical properties of DN hydrogels. The hydrogels with nearly 90 wt% water had achieved fracture strain of about 2400% and a maximum compression stress of 35.2 MPa. Moreover, we also observed the hydrogels could quickly recover to their initial shapes after the deformation force was removed. By introducing a molecular stent, we anticipated that a large number of neutral synthetic polymers would be utilized to form the first network in DN hydrogels, significantly expanding the scope of the DN hydrogel concept.

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1. Introduction

A hydrogel that could absorb water or biological fluid but not dissolve is a three-dimensional (3D) network polymer cross-linked by covalent or strong physical bonds [1]. The hydrogel as a "soft-and-wet" material has some unique properties, including biocompatibility, responsiveness to various kinds of stimuli, low surface friction and friendly to environment [2,3]. As a result, the hydrogels have attracted much attention as novel functional materials in the applications of medical and industrial fields, such as 3D printing for soft tissues [4], vehicles for drug delivery [5], extracellular matrices for biological studies [6], actuators and sensors [7], and so on. However, the fetal weakness of hydrogels prepared by single polymers is their poor mechanical behavior. For example, an alginate hydrogel would rupture when stretched to about 1.2 times its original length [8].

A large number of investigations are devoted to improve the mechanical properties of hydrogels, such as double network (DN) hydrogels [9], nanocomposite hydrogels [10], slide-ring hydrogels [11], triblock copolymers hydrogels [12], hydrophobic modified hydrogels [13], tetra-PEG gels [14] and macromolecular microsphere composite (MMC) hydrogels [15]. Among them, the DN hydrogel concept was firstly developed by Gong and co-workers, and would be an effective approach to enhance the mechanical properties of hydrogels with high strength and toughness [9,16]. Usually the DN hydrogels are prepared by a two-step sequential free-radical polymerization process. The strong polyelectrolytes are used as the

* Corresponding authors.
E-mail addresses: xyren_bio@163.com (X.Y. Ren), ghgao@ccut.edu.cn (G.H. Gao).

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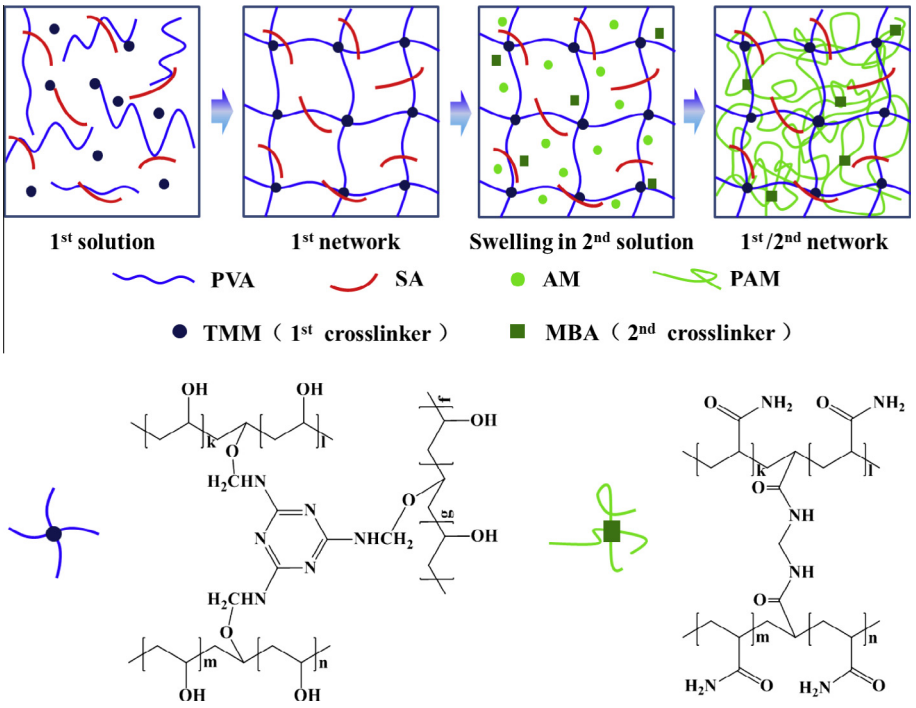

Fig. 1. Schematic illustration of the preparation of PVA-SA/PAM DN hydrogels using a neutral synthetic polymer PVA as the first network, PAM as the second network and SA as a molecular stent. Due to an extra ionic osmotic pressure provided by SA, the neutral first-network polymer can absorb a large amount of the second-network aqueous media.

Table 1
The components of the samples in semi-IPN and DN hydrogels.

First network	Second network	PVA mole concentrations (mol/L)	TMM mole fractions (mol%)	AM mole concentrations (mol/L)	MBA mole fractions (mol%)	SA (wt%)
PVA-1.5-6-SA	PAM-2.5-0.05	1.5	6	2.5	0.05	0
PVA-1.5-6-SA	PAM-2.5-0.05	1.5	6	2.5	0.05	10
PVA-1.5-6-SA	PAM-2.5-0.05	1.5	6	2.5	0.05	20
PVA-1.5-6-SA	PAM-2.5-0.05	1.5	6	2.5	0.05	30
PVA-1-6-SA	PAM-2.5-0.005	1	6	2.5	0.005	30
PVA-1-6-SA	PAM-2.5-0.01	1	6	2.5	0.01	30
PVA-1-6-SA	PAM-2.5-0.03	1	6	2.5	0.03	30
PVA-1-6-SA	PAM-2.5-0.07	1	6	2.5	0.07	30
PVA-1-6-SA	PAM-2.5-0.1	1	6	2.5	0.1	30
PVA-1-6-SA	PAM-2.5-0.2	1	6	2.5	0.2	30
PVA-1-6-SA	PAM-1.5-0.03	1	6	1.5	0.03	30
PVA-1-6-SA	PAM-2.5-0.03	1	6	2.5	0.03	30
PVA-1-6-SA	PAM-3-0.03	1	6	3	0.03	30
PVA-1-6-SA	PAM-3.5-0.03	1	6	3.5	0.03	30
PVA-1.5-5-SA	PAM-2.5-0.03	1.5	5	2.5	0.03	20
PVA-1.5-6-SA	PAM-2.5-0.03	1.5	6	2.5	0.03	20
PVA-1.5-7-SA	PAM-2.5-0.03	1.5	7	2.5	0.03	20
PVA-1.5-8-SA	PAM-2.5-0.03	1.5	8	2.5	0.03	20

rigid and brittle first network via highly covalently cross-linking. Then, the neutral monomers would infiltrate into the first network hydrogels and form loosely cross-linked, soft and ductile, second network hydrogels [9,17]. In contrast, the neutral hydrogels as the first network exhibited poorer swelling ability in the second network monomers and the resulting DN hydrogels had both poor strength and poor extensibility. Subsequently, Gong and coworkers developed a molecular stent method to use neutral or weak polyelectrolytes as the first network in the DN hydrogels [18]. However, the limitation for neutral synthetic polymer to form first network hydrogels in DN hydrogels cannot be solved.

Poly(vinyl alcohol) (PVA) is a neutral synthetic polymer with a simple chemical structure and has an excellent history of biomedical applications [19,20], specifically in the form of hydrogels. Oka et al. found no inflammatory or degenerative changes in the articular cartilage or synovial membrane by using the artificial PVA cartilages after 8–52 weeks [21].

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