Abstract

Oriented hydrogels have been widely studied in fields of soft-biomaterials due to their anisotropic properties possibly applicable in biomedics, optics, and electronics. According to the literatures, network structures of the hydrogels were oriented by various factors such as self-assembly under flow, mechanical elongation, solvent-cast, etc. However, no report on one-directional swelling of the hydrogels has been available in spite of ultimate targets for the anisotropic hydrogels. Molecularly-oriented hydrogels of sacran, which is a supergiant liquid crystalline polysaccharide extracted from *Aphanothece sacrum* biomaterials, showing ultra-high anisotropy of swelling is successfully prepared by two-step chemical cross-linking. Divinylsulfone (DVS) works as a chemical cross-linker of sacran chains in a dilute aqueous solution to form hydrogels, but some of the added DVS remains in the hydrogel without cross-linking. The remaining DVS cross-links further with the pre-formed networks of sacran chains in liquid crystalline state during slow drying to produce in-plane oriented xerogels. The xerogels show heterogeneous anisotropy in the successive swellings steps; the linear swelling ratio in the thickness direction is 10,000-40,000 fold higher than that in the width direction, due to the molecular orientation of the sacran hydrogels. X-ray diffraction imaging of the hydrogels reveal not only the orientation of the xerogel films but also the unusual orientation of water molecules binding to sacran networks in the hydrogel state. The oriented hydrogels show various anisotropies such as mechanical properties and stimuli-responsiveness.

Keywords: anisotropy, hydrogels, polysaccharide, liquid crystals, in-plane orientation