

Investigating the in-situ structure-property of carrageenan gels and its mixtures using large deformation via stretching and small angle neutron scattering

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The development of hydrogels is important in a wide range of applications in biotechnology and bioengineering. In particular, hydrogels with high flexibility and elasticity and excellent mechanical properties are very attractive in many biomedical applications, for example, in artificial muscles and cartilages. However, in most cases, hydrogels with excellent mechanical properties have been achieved using synthetic polymers whose network structure and cross-linking can be controlled and designed. On the other hand, natural biopolymers such as polysaccharides have attracted much attention over the years for their high biocompatibility, biodegradability, and environmentally friendly materials made from renewable resources.

K-carrageenan is a linear polysaccharide known for its impressive gelling properties, particularly through physical crosslinking and the formation of double helices in the presence of cations. Its biocompatibility and biodegradability make it a promising biopolymer for hydrogel applications. However, traditional carrageenan gels often exhibit either brittleness or weakness, limiting their practical uses. Initially, we propose to utilize the mixtures of carrageenan hydrogels to improve the mechanical property; however, it was found that the tuning the crosslinking points of the carrageenan hydrogels is more effective. Furthermore, to implement in-situ stretching/SANS experiments, the mechanical property of the gels is very important. Thus, to enhance the mechanical property of carrageenan hydrogels, we introduce a dual crosslinking strategy that combines both chemical and physical crosslinking.

Carrageenan forms a network by aggregation of molecular chains in a gel, but large deformation causes the aggregation structure to break down, leading to macroscopic rupture. In addition, to understand the toughening mechanism of dual

crosslinked carrageenan gels, the nanostructure under deformation will be investigated by in-situ SANS measurement.

Figure 1 shows the intensity profile after circular averaging for carrageenan gels without and with deformation. For carrageenan gel without deformation (blue), a broad shoulder appears at about Q 0.004 \AA^{-1} . This shoulder is attributed to the presence of aggregates composed of double helices. Furthermore, the intensity demonstrates an increase with decreasing q based on the power law $I(Q)=Q^{-3}$ suggesting the fractal dimension of the aggregates.

On the one hand, the intensity around the broad shoulder decreases with stretching. It is considered that stretching caused the reorientation and realignment of the aggregates parallel to the stretching direction. Thus, this decrease may be ascribed to the reorientation of the aggregates.

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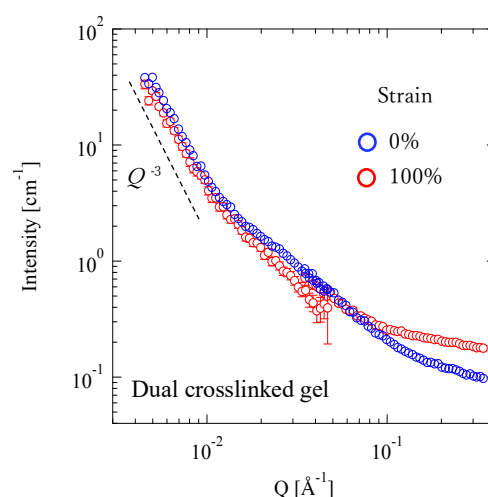


Fig. 1. 1D intensity profile of dual crosslinked carrageenan gel with (red) and without (blue) stretching.