Elaborating the origin of tough carrageenan gels based on the dynamic of the aggregates using neutron spin echo

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Stretchable and tough hydrogels are important materials for the development of several applicatons most especially in biomedical field. These hydrogels have the capability to mimic the properties of biological tissue such as muscle, cartilage, etc. Moreover, the development of hydrogels opens plethora of applications in advanced biotechnological applications including smart wearables, among other. However, in most cases, tough hydrogels are only achieved by using synthetic polymers where the network structure and crosslinks can be controlled and designed. Meanwhile, the idea of a tough biopolymer-based hydrogels is very attractive given the high biocompatibility of the biomaterials. However, the attempt to develop a tough hydrogel from polysaccharide alone without any addition of synthetic polymers often failed due to the difficulty in controlling the network structure and predicting the dynamics of the network.

Recently, we were able to develop a tough and stretchable polysaccharide-based hydrogels from carrageenan. However, a full understanding of the spatial and temporal characteristics of a dual crosslinked carrageenan network is still lacking, which is crucial in the development of the tough and stretchable carrageenan gels.

Figure 1 (top) presents to the 1D-SANS scattering profile of tough carrageenan gels prepared with different types of salts, namely KCl and $ZrOCl_2$. Gels prepared with pure KCl formed soft with breaking stress. Meanwhile, the gel prepared with 50%KCl:50%$ZrOCl_2$ produced a stiff and tough biobased hydrogel. Based on the SANS results, it is proposed that the increased stiffness of the gel is attributed to the enhanced aggregation of the carrageenan double helices. Thus, it is believed that a hierarchical structure of double helix bundles was established in the hydrogel, most especially gels with $ZrOCl_2$ [1].

Meanwhile, dynamics studies by neutron spin echo (bottom) suggest almost no mobility was observed for either sample at a very short time scale (t<10 ns) due to the highly restricted aggregates, but significant difference was seen by pulsed NMR. This indicates the formation of aggregates with highly restricted molecular mobility, consistent with the structural analysis by SAXS and SANS.

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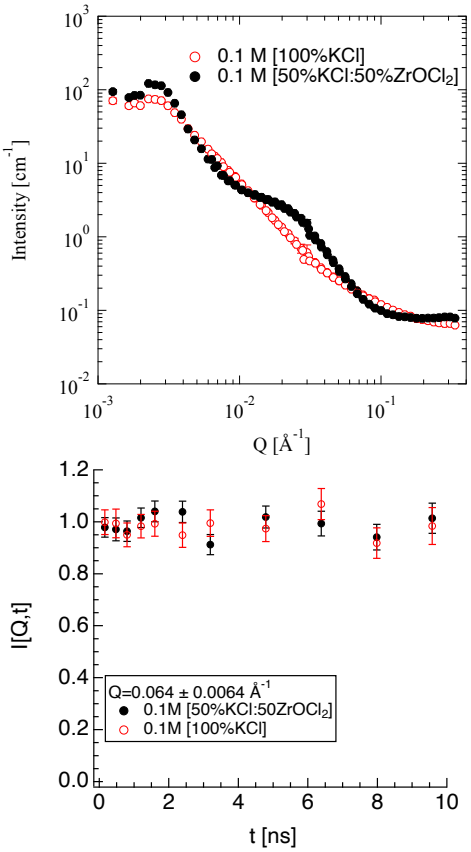

Fig. 1. (top) 1D small angle neutron scattering profile obtained at SANS-U. (bottom) The intermediate scattering function I(Q,t) of the hydrogels obtained by the neutron spin echo instrument, iNSE at C2-3-1 at Japan Research Reactor-3 (JRR-3).