Gloiopeltis furcata由来フノラン：抽出、化学構造決定及びゲル化の速度論

[課程博士・論文博士共通]

博士学位論文内容要旨
Abstract of Dissertation

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論文題目 Title	Funoran from *Gloiopeltis furcata*: Extraction, Chemical Characterization, Gelation and Kinetic Mechanism		

Funoran, a sulfated polysaccharide derived from red algae (Gloiopeltis species), is a heterogeneous polysaccharide consisting of the major repeating unit of β-D-galactose-6-sulfate and 3,6-anhydro-α-L-galactose. Funoran has gained significant attention in diverse industries due to its non-toxic and biocompatible properties. Its adhesive qualities are particularly valuable in papermaking and textile printing, where it acts as a binder, improving the strength and durability of products. The food industry values funoran for its thickening and stabilizing abilities, which enhance the texture and consistency of food products like dairy desserts, sauces, and dressings. The application of funoran polysaccharide can be extended in food, medical and other industry fields through understanding its physical and chemical properties.

The gelation process is crucial for achieving the desired texture in many food products. The gelation mechanism of funoran depends on the chemical structure of the polysaccharide and the extrinsic like temperature, ion species and so on. The chemical structure of funoran affected many factors such as algae species, growth geography, harvest season, and extraction and purification methods.

The gelation process is described as a dynamic change from the solution phase to the gel phase. Studying the kinetics of the gelation of funoran is important for providing insights into how the gel forms and develops over time. Rheological measurement and the signal intensities in ^{1}H NMR are effective tools for observing and classifying structural changes which occur during the time-dependent gelation process. By understanding the funoran gelation kinetics, scientists and food technologists can optimize the gelation process to achieve the desired texture and quality of food products. Therefore, this study focused on two experiments;

In the first experiment, two extraction strategies were used to obtain funoran from *Gloiopeltis furcata* with different physicochemical properties. Strategy 1 involved direct hot water extraction, yielding two fractions. Strategy 2 involved cold water purification followed by hot water extraction, resulting in two additional fractions. All fractions contained β-D-galactose-6-sulfate and 3,6-anhydro-α-L-galactose units as the backbone. The fractional difference in strategy 2 was clearly observed in which the high molecular weight formed a highly viscous and cloudy solution with low solubility, while the low molecular weight was highly soluble and formed a clear and transparent solution. All funoran fractions in a 1.5% aqueous solution (w/w) exhibited shear thinning behavior at 20 ℃. Cooling-induced gel formation, observed by a sharp increase in complex modulus $|G^*|$, and reheating showed thermal hysteresis with confirmed gelation using micro-DSC measurements. These findings demonstrate the impact of extraction strategies on funoran properties, including solubility, rheological behavior, and gelation characteristics.

In the second experiment, the rheological measurements were conducted with aging time. The gels quenched at lower temperatures (2 °C) exhibited higher storage modulus, gel strength and faster gelation rate than those quenched at higher temperatures (20 °C). From the rheological measurements, the gelation rate of the funoran system showed strong time dependence during the aging process at different temperatures. The rheological data was successfully fitted with the model of a combined reaction of the 2nd order at the nucleation step (coils interaction) and the 1st order at the crosslinking progress (hydrogen bond formation) with a time course that affects the aggregation length. Consequently, during the aging time, higher temperature (20 °C) required longer helices for a stable association to keep crosslinking region, resulting in a slow rate compared to numerous shorter helices at lower temperatures (2 °C). From the ^{1}H NMR results, it was found that the change in signal intensities was consistent with the macroscopic gelation process as observed in the rheological measurement where a slow decay in the signal intensities was observed at 2 °C but the gradual decay was observed at high temperature (20 °C) with aging time.

In summary, this study aims to provide a simple way to isolate funoran fractions with significantly different solvation and gel-forming ability from *G. furcata*, which should be useful for promoting the industrial isolation of funoran with different properties. The gelation process in funoran was influenced by temperature and time. From the successful theoretical fitting model with the rheological measurements and ^{1}H NMR results, the funoran system's gelation rate showed strong time dependency at different temperatures. This knowledge can provide insights for optimizing the gel properties of funoran in food applications.