

Rheology of highly elastic iota-carrageenan/kappa-carrageenan/xanthan/konjac glucomannan gels



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ABSTRACT

Mixed gels containing κ -carrageenan, ι -carrageenan, konjac glucomannan and xanthan were investigated. In binary mixtures, the only interactions identified from differential scanning calorimetry were between konjac glucomannan and κ -carrageenan or xanthan. The effect of konjac addition on κ -carrageenan- ι -carrageenan mixed gels was similar to that on κ -carrageenan gels, namely, an initial increase of the elastic modulus was observed up to a certain stoichiometric ratio, followed by a plateau of the elastic modulus and a strong increase of the fracture strain when konjac was added in excess. Addition of xanthan led to maxima and minima of the elastic modulus and fracture strain and stress. Interestingly, characteristic features of the elastic modulus appeared at the same concentration of xanthan added to kappa-carrageenan/konjac glucomannan mixtures with or without iota-carrageenan, indicating that xanthan is present in the kappa-carrageenan/konjac phase and absent from the iota-carrageenan phase. Characteristic features of the fracture properties, however, were shifted to higher concentrations of xanthan when iota-carrageenan was added. We tentatively conclude that xanthan chains that are concentrated in the kappa-carrageenan/konjac phase under quiescent conditions achieve homogeneous distribution under large mechanical deformation.

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1. Introduction

As described by Nishinari et al. (2008), the importance of texture in Japanese culture is underlined by the much larger number of texture terms and their frequency of use in Japanese compared with other languages. With the accelerating globalization of the food market, it is not out of the question that English speakers will in the future be equally familiar with Japanese terms like *mochi-mochi*, which refers to high chewiness or rubbery texture, as with the Italian *al dente* or the French *au gratin*.

Chewiness is not always a desirable textural feature of foods. A leathery texture of apples is clearly undesirable (Bourne, 1986), while it is usually undesirable for bread (Gray & BeMiller, 2003) and

often undesirable for meat and fish (Brauer, 1993; Sveinsdottir et al., 2009; Sveinsdottir, Martinsdottir, Hyldig, & Sigurgisladottir, 2010). However, several popular Japanese foods are characterized by very high elasticity and an almost “rubbery” texture that requires prolonged mastication. Obvious examples are Oden sausages, which consist of elastic gum-covered meat; Warabimochi, a dessert gel based on plant polysaccharides; and mochi, a glutinous rice cake, traditionally eaten at New Year celebrations. In addition, some Japanese dessert jellies have a very elastic texture that leads to prolonged mastication times. The high elasticity of several such model gels has been identified as a major factor contributing to effort of eating (Hayakawa et al., 2014), which in elderly individuals that suffer from deficient mastication strategies can lead to danger of choking and/or dysphagia. In spite of this designation of highly elastic foods, very soft and rubbery dessert jellies have been suggested as possible foods for elderly with mastication problems (Funami, Ishihara, Nakauma, Kohyama, & Nishinari, 2012).

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The carrageenan family encompasses structurally similar poly-anionic galactans derived from red algae. The industrially most important carrageenan is the κ form, rendering it one of the most popular polysaccharides in the food industry (Piculell, 2006). At fixed total carrageenan concentrations, κ -carrageenan mixtures with its related ι form are characterized by lower elastic moduli and a higher fracture strain, resulting in a peak of the fracture stress around equal mixing ratios (Brenner, Tuvikene, Parker, Matsukawa, & Nishinari, 2014; Rochas, Rinaudo, & Landry, 1989). Mixtures of κ -carrageenan with the plant polysaccharides locust bean gum and konjac glucomannan can show both a higher elastic modulus and a higher fracture stress than pure κ -carrageenan, depending on the mixing ratio (Brenner & Nishinari, 2014; Brenner, Wang, Achayuthakan, Nakajima, & Nishinari, 2013). Xanthan, a cellulosic-backbone microbial polysaccharide, has been shown to phase-separate from λ -carrageenan and other negatively charged polysaccharides (Boyd et al., 2009). In the presence of konjac glucomannan it is known to form synergistic gels (e.g., Annable, Williams, & Nishinari, 1994).

In the present investigation, we map the rheological properties of polysaccharide mixtures known to produce highly elastic gels, based on κ -carrageenan in the presence of ι -carrageenan, xanthan and konjac glucomannan. The results are important for enriching the texture range available for elastic polysaccharide containing foods, especially jellies.

2. Materials and methods

2.1. Chemical characterization

All polysaccharide powders were a gift from San Ei Gen F.F.I. (Osaka, Japan) and were used without further purification. KCl was purchased from Sigma–Aldrich. The contents of the κ -carrageenan (KC, SAN SUPPORT® G-16) and ι -carrageenan (IC, SAN SUPPORT® G-17) powders were reported in a previous publication, with impurities of 19.5 mol% ι -carrageenan and 1.7 mol% κ -carrageenan in KC and IC, respectively (Brenner, Tuvikene, et al., 2014).

Size exclusion chromatography was performed as described previously (Brenner, Tuvikene, et al., 2014), where the peak molecular weights of our KC (1860 kDa) and IC (1435 kDa) samples were given. The size exclusion chromatograms of xanthan (XAN, VIS TOP® D-3000-C) and konjac glucomannan (KGM, VIS TOP® D-2134) are given in Fig. 1. The peak molecular weights of XAN and KGM are 7030 and 2200 kDa, respectively. We note that the molecular weight for XAN is higher than that of the largest standard used, and contains therefore considerable uncertainty. The peaks for nitrate and chloride ions are indicated in the figure. The peak to the left of the Cl^- peak corresponds to some small inorganic compound that we were unable to identify.

Inductively coupled plasma measurements were carried out as previously reported (Brenner, Tuvikene, et al., 2014), and the results are given in Table 1.

Monosaccharide and side-group contents of XAN and KGM are given in Table 2. Glucose and mannose contents were obtained by hydrolysis of the polysaccharides in 2 M H_2SO_4 at 110 °C for 60 min (KGM) or 120 min (XAN) followed by neutralization with 1 M NaOH; the monosaccharides were quantified by HPAEC-PAD using the CarboPac MA1® column at 35 °C (Quemener & Lahaye, 1998). Uronic acids were quantified by the 3-phenylphenol colorimetric method described by Filisetti-Cozzi and Carpita (1991). D-Glucuronic acid was used as a standard. The determination of acetate content was carried out by mild alkaline treatment of the polysaccharide (0.5%) in 0.02 M NaOH for 24 h at 25 °C. The titration of

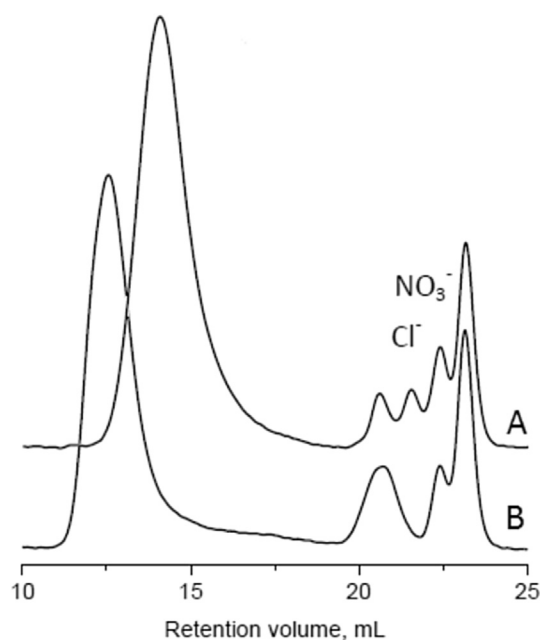


Fig. 1. Size exclusion chromatograms of (A) KGM and (B) XAN obtained at 60 °C.

Table 1

Weight content (%) of select elements, moisture and total polysaccharide (without counterions) in polysaccharide powders. Content is per dry powder weight.

	IC	KC	XAN	KGM
Na	0.84	0.32	0.91	0.25
K	3.3	9.1	3.4	3.6
Ca	3.3	0.88	0.07	0.17
Mg	0.21	0.18	0.08	0.10
S	13.1	7.7	–	–
Moisture	10.3	8.7	10.5	8.4
Total polys.	80.2	71.0	80.0	87.6

the remaining NaOH was performed with 0.01 M HCl using phenolphthalein as an indicator. The pyruvate content of xanthan was determined from the acetate/pyruvate ratio as observed in the ^1H NMR spectrum (Rinaudo, Milas, Lambert, & Vincendon, 1983). ^1H NMR spectroscopic analyses were performed on a Bruker AVANCE III spectrometer operating at 800 MHz. The ^1H NMR spectra from 0.05% polysaccharide solutions in D_2O (w/w) were obtained at 70 °C and 64 transients were collected. The molecular weight of the polysaccharide samples was reduced prior to the NMR measurement by sonicating in a water-bath at 25 °C for 100 min with an ultrasound homogenizer (Bandelin Sonopuls UW2070, 70 W, operating at 80%), followed by centrifugation and freeze-drying.

Table 2

Content of acetate (based on $M_w = 59$ Da) and pyruvate (based on $M_w = 87$ Da) per dry powder weight, along with M/G (mannose/glucose) and M/GA (mannose/glucuronic acid) ratios in XAN and KGM.

	XAN	KGM
Acetate	5.2%	1.7%
Pyruvate	5.3%	–
M/G ratio	0.88	2.03
M/GA ratio	2.19	–

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