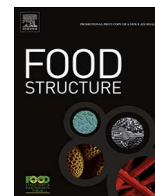




Contents lists available at ScienceDirect

## Food Structure

journal homepage: [www.elsevier.com/locate/foostr](http://www.elsevier.com/locate/foostr)

# Effect of alcohols on gellan gum gel structure: Bridging the molecular level and the three-dimensional network

Mattia Cassanelli\*, Ian Norton, Tom Mills

School of Chemical Engineering, University of Birmingham, Edgbaston, Birmingham, B15 2TT, UK

## ARTICLE INFO

## Keywords:

Gellan gum  
Solvent quality  
Alcogel

## ABSTRACT

This work focuses on the understanding of the interaction of alcohols with gel systems during solvent exchange, following the gel formation. A method of widening the possible alcohol contents in formulations is proposed, as most hydrocolloids have a low tolerance of high alcohol concentrations and in some gelation is completely prevented.

Once the CPKelco LA (low-acyl or deacylated) gellan gum gel was produced, different alcoholic solvents (ethanol, 1-propanol, 2-propanol) were used to remove water from the material and replace it, investigating the effect on the gel network as a function of the alcohol chain length.

It is the first time that a research paper considers the high-alcohol/gellan gum systems at both the molecular and macroscopic scales, proposing the link between them. Specifically, the interaction of the alcoholic solvents with both the polymer chains and three-dimensional network was evaluated by characterising the physical and mechanical gel properties throughout the alcohol treatment.

From this study, the solvent effect on the gelling agent is evident, leading to structure shrinkage and distortion due to a high-induced stress on the gellan gum network. Gradual addition of the alcoholic solution was found to considerably reduce this behaviour due to the slower solvent exchange.

As evidence, and to further validate this study on LA gellan gum, both k-carrageenan and gelatin alcogels were investigated, since their gelation mechanism and molecular configuration are respectively similar and different to LA gellan gum. It was found that k-carrageenan reproduced the LA gellan results, unlike gelatin.

## 1. Introduction

The use of alcohols in combination with gelling agents has been increasing in different industrial fields. In the food industry, hydrocolloids are often used with ethanol in both food and beverage products to provide functional properties (Chaudhary, Small, & Kasapis, 2013; Imeson, 2012), such as the system homogeneity and stability over time (Chalupa, King, & Giampetro, 1997; Imeson, 2012), the viscosity enhancement (Khan, Park, & Kwon, 2007) and the soft tribology improvement (Mills, Koay, & Norton, 2013). Moreover, alcohols might be found in drying processes, such as in the supercritical-fluid drying of both gel systems (Scherer, 1990; Tkalec, Knez, & Novak, 2015; Ubeyitogullari & Ciftci, 2016) and entire food products (Brown, Fryer, Norton, Bakalis, & Bridson, 2008; Garcia-Gonzalez et al., 2007).

In the biomedical sector, alcohols may be used to dry and sterilise hydrogels (Perrut, 2012; Shearer, Ellis, Perera, & Chaudhuri, 2006) and to prepare them for microscopy characterisation (Allan-Wojtas, Hansen, & Paulson, 2008). Furthermore, in tissue engineering ethanol is

widely used in combination with biopolymers during scaffold preparation (Dai, Ronholm, Tian, Sethi, & Cao, 2016) and for the decellularisation process (Sawada, Terada, Yamaoka, Kitamura, & Fujisato, 2008).

Some hydrocolloids (e.g pectin and guar gum) can still form a gel network if alcohols are added to the hot solution during the preparation stage (Oakenfull & Scott, 1984; Phillips & Williams, 2000; Tkalec et al., 2015). In this case, the used term is alcogel (Tkalec et al., 2015). However, the alcohol percentage depends on the specific gelling agent and can be limited. For instance, hydroxypropyl cellulose (HPC) is soluble in aqueous solution with an ethanol concentration around 50%, while xanthan gum water-solution can contain up to 60% in ethanol (Phillips & Williams, 2000). Furthermore, the ethanol addition can affect some gel properties, like transparency, and promote gelation at lower temperatures (Yamanaka et al., 2000).

In this context, a common gelling agent used in the food industry is LA (low-acyl or deacylated) gellan gum (Khan et al., 2007; Morris, Nishinari, & Rinaudo, 2012), which is a microbial polysaccharide,

\* Corresponding author.

E-mail address: [mxc446@student.bham.ac.uk](mailto:mxc446@student.bham.ac.uk) (M. Cassanelli).<http://dx.doi.org/10.1016/j.foostr.2017.09.002>

Received 15 May 2017; Received in revised form 5 September 2017; Accepted 18 September 2017

2213-3291/ © 2017 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

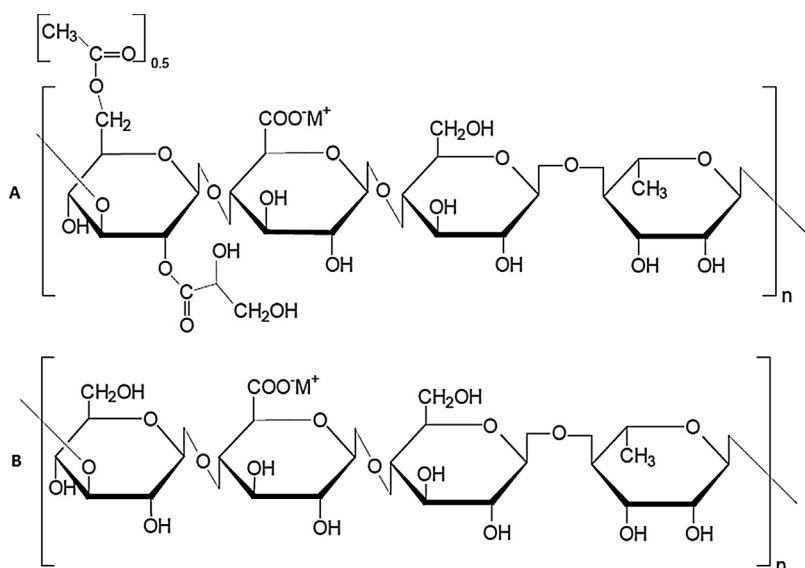


Fig. 1. HA gellan gum (A) and LA gellan gum (B) polymeric chains (adapted (Mahdi, Conway, & Smith, 2015)).

produced by the microorganism *Sphingomonas elodea* (previously identified as *Pseudomonas elodea*) in a fermentation process. The primary structure is a tetrasaccharide unit composed of glucuronic acid, rhamnose and glucose (Phillips & Williams, 2000), with a molecular weight range between 100 and 200 kDa (CPKelco, 2007; Kirchmayer, Steinhoff, Warren, Clark, & in het Panhuis, 2014). In the adapted Fig. 1 (Mahdi, Conway, & Smith, 2015), both the high-acyl (HA) and deacylated gellan gum chains are shown.

The two gellan gum types can be blended to provide synergistic properties to the system, especially in terms of mechanical properties (Phillips & Williams, 2000), whose ratio and total solid content depends on the specific application.

Morris, et al. (2012) reported that the gelation mechanism for gellan gum starts by formation of double helices, and, afterwards, the ion-induced association of the double helices leads to junction zone formation. In other words, the gellan network consists predominantly of flexible, disordered chains, with few ordered junction zones between the helices. For these zones, stabilising forces such as hydrogen bonds, electrostatic forces, hydrophobic interactions, Van der Waals attractions and molecular entanglement are defined by the solvent conditions and polymer structure (Hui, 2006). It is reported that low acyl gellan gum requires cations, acid, soluble solids or combinations of these additives (CPKelco, 2007; Morris et al., 2012; Norton, Cox, & Spyropoulos, 2011).

Since gellan gum is not soluble in ethanol (Bajaj, Survase, Saudagar, & Singhal, 2007), yet the product formulation may contain both the ingredients, this work proposes a method to widen the alcohol content in gelling agent systems. LA gellan gum was chosen as a gel model system, representing other hydrocolloids with similar gelation mechanism (Aguilera & Stanley, 1999). The characterisation at the molecular scale was performed by mDSC and FTIR, as Sudhamani, Prasad, and Udaya Sankar (2003) reported for the gellan gum gels without alcohols, whereas the mechanical properties were investigated by texture analysis.

The alcohol-hydrocolloid interaction was assessed when the gel was already produced. Particularly, quiescent gels (Morris et al., 2012) were evaluated to study the effect on the molecular/network structures. However, this study is also applicable to smaller aggregates, e.g. gel particle suspensions (Banerjee & Bhattacharya, 2012; Norton, Jarvis, & Foster, 1999).

## 2. Material and methods

### 2.1. Gel preparation and solvent treatment

Double distilled water, obtained by a water still system (Aquatron A400D, Stuart, UK), was heated up to 85 °C and then the low acyl gellan gum powder (Kelcogel F, CPKelco, UK) was slowly added to avoid clump formation. The polymer concentration was 2% w/w in order to have a stable quiescent gel block, yet not too dense. To ensure a complete hydration, the solution was stirred for two hours at constant temperature.

No salts were added to strengthen the gel (Phillips & Williams, 2000) in order to avoid introducing a further variable to the system, and potentially affecting the results on the solvent interaction with the gel network in both mechanical and chemical properties.

The solutions were poured into sample moulds (13.5 mm in diameter and 65 mm in height), which were covered with a plastic film to prevent evaporation. A cooling rate of around 0.5 °C/min down to room temperature (20 °C ± 1 °C) was recorded. This specific cooling rate was kept constant during all the experiments to minimise changes in the gel structure, as Nitta, Yoshimura, and Nishinari (2014) suggested.

After the gel setting, the moulds were stored at room temperature (20 °C ± 1 °C) for 24 h.

Afterwards, the obtained gel samples had dimensions of 13.5 mm in diameter and 10 mm in height.

Similarly, 2% w/w gelatin (from porcine skin, Sigma-Aldrich, UK) and 2% w/w k-carrageenan (Sigma-Aldrich, UK) were prepared to compare the solvent quality with LA gellan gum.

Once the gels were formed, different alcohols were separately used to assess their effect on the gel properties. Ethanol (EtOH), 1-Propanol (1-PrOH) and 2-Propanol/Isopropanol (2-PrOH) (AnalaR NORMAPUR, VWR, UK) were used as pure solvents or diluted in different concentrations with distilled water to perform a gradual alcohol treatment. In this specific case, solutions at 25, 50 and 80 wt% were prepared. When a gradual treatment was applied, the gel samples were left, stepwise, in the alcoholic solution for 6 h. Finally, the treated gels were submerged in the pure solvent. The last step of the gradual treatment was 24-h long. On the other hand, if the treatment was not gradual, the samples were directly left for 24 h in the specific alcoholic solution/pure alcohol.

### 2.2. Mechanical and shrinkage measurements

Mechanical properties were evaluated by analysing the material

Download English Version:

<https://daneshyari.com/en/article/6489506>

Download Persian Version:

<https://daneshyari.com/article/6489506>

[Daneshyari.com](https://daneshyari.com)