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# Mechanical properties of gelatin-rich micro-particles

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**Abstract**

Effect of chemical composition and applied strain on mechanical properties of gelled gelatin-rich micro-particles resulting from phase separated gelatin/pullulan mixtures has been investigated. The mechanical properties of micro-particles (20–120 μm) were measured using a micromanipulation technique. The compress–release tests revealed that at a low deformation (up to 0% strain) particles are fully elastic with Young's modulus proportional to the concentration of gelatin and at the higher deformation (up to 50–80%) particles are visco-elastic. Even at very high load resulting in 50–60% deformation, no fracture of particles was observed and after the load was removed, particles recovered to a fully spherical shape. The visco-elastic behaviour was investigated by a stress-relaxation method, where force relaxation at constant deformation was measured as a function of time. The experimental results were analysed using a standard liner model of visco-elastic solids and the parameters of this model were related to the composition of gelled particles.


*Keywords:* Gelled micro-particles; Gelatin-rich phase; Mechanical properties; Standard linear model

## 1. Introduction

Phase separated biopolymers mixtures in which one or both phases gel during processing are frequently used in different types of manufactured food products. The microstructure of such products has a strong effect on texture and flavour and it can be controlled by selection of composition of the mixtures and the processing parameters such as temperature, cooling and shear rates (Wolf, Scirocco, Frith, & Norton, 2000). Processing of phase separated mixtures in which gelation occurs can be seen as a two-step process. In the first step, carried out at the temperature above the gelling point of either of the polymers, the required structure is formed and in the second step, the temperature is reduced at the prescribed rate to well below the gelling point (other process parameters might also be adjusted) trapping the structure (Wolf, Frith, Singleton, Tassieri, & Norton, 2001). Understanding of the effect of the composition, temperature and the shear rate on the rheological properties and interfacial tension in such mixtures above the melting point is essential when the structure is created (Ding, Pacek, Frith, Norton, & Wolf, 2005), whereas the mechanical properties of gelled particles are important in the step used to kinetically trap the microstructure which then controls the perception of the taste of the food products (Foegeding, 2005).

Particulate gels are of growing interest in the food industry as the consumer wishes to have good quality enjoyable and convenient foods but with a healthier formulation. The use of gel particles to mimic fat droplets is one such route, however, if this is to be achieved then, we must be capable of designing the material properties of the gelled particles to be the same as the material properties of the emulsion droplets they replace (Norton, Fryer, & Moore, 2006).

The mechanical properties of a single component gelatin gels have been extensively investigated and it has been found that they depend on the type (bloom number) and the concentration of gelatin, presence of additives such as salts and/or other proteins, pH of gelatin solution prior to gelation and temperature (Bot, van Amerongen, Groot, Hoekstra, & Agterof, 1996; Gomez-Guillen et al., 2002).

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**Nomenclature**

| | | | |
|---|---|---|---|
| $a$ | constant | $R$ | initial radius of gelled particles (m) |
| $b$ | constant | $\varepsilon$ | pseudo strain, $(h/R)$ |
| $c$ | constant | $\varepsilon_\infty$ | $\varepsilon(t \to \infty)$ |
| $D$ | deformation parameter $(h/2R)$ | $\eta$ | viscous modulus in Kelvin–Voigt element (Pas) |
| $E$ | Young's modulus (Pa) | $\lambda'$ | relaxation time (s) |
| $E'$ | Young's modulus in Kelvin–Voigt element (Pa) | $v$ | Poisson's ratio |
| $E_\infty$ | $E(t \to \infty)$ (Pa) | $\sigma$ | pseudo stress, $F/\pi R^2$ (Pa) |
| $F$ | force (N or mN) | $\sigma_0$ | $\sigma(t = 0)$ (Pa) |
| $h$ | displacement (m) | $\sigma_i$ | constant in Eq. (4) |
| $k_1$, $k_2$ | constants in Eq. (6) | | |

Djabourov (1991) reported strong effect of ageing time and temperature on physical/mechanical properties of several types of gels. It has been also reported that tensile strength and fracture strain strongly depend on a strain/deformation rate with the fracture strain increasing with the gelatin concentration (McEvoy, Ross-Murphy, & Clark, 1985a). In general, it has been accepted that at small deformation gelatin gels are linearly elastic and relation between stress and strain is fully characterised by Young's modulus but at large deformation the relation between strain and stress is nonlinear and more complex models are necessary to describe the stress–strain relation.

The mechanical properties of composite gels of phase separated structures (approximately spherical inclusions of dispersed phase embedded in matrix phase) were also investigated. McEvoy, Ross-Murphy, and Clark (1985b) investigated tensile strength of composite gelatin/agarose gels at large deformation and showed that the shear or Young's modulus of such gels can be related to the moduli of the component gels. Plucknett, Normand, Pomfret, and Ferdinando (2000) found that the failure of mixed gelatin/maltodextrin gels is mainly caused by de-bonding between the dispersed particles (maltodextrin rich gel) and the continuous phase (gelatin-rich gel). Recently, Rizzieri, Baker, and Donald (2003) investigated the large strain deformation and failure in the same mixed gels system and employed an environmental scanning electron microscope (ESEM) for detailed analysis of the changes of the structure of the composite gels during a tensile test. They observed ductile behaviour attributed to the debonding at the interface between maltodextrin particles and continuous gelatin-rich matrix phase as well as to the deformation and fracture of the included maltodextrin phase.

Whilst the mechanical properties of a single component gelatin gels as well as composite gels containing gelatin were extensively investigated, there is practically no information in the open literature on mechanical properties of gelatin-rich phase resulting from gelation of gelatin-rich droplets in phase separated mixtures. This work fills this gap and reports the results of the investigation of the effect of the composition and strain on the mechanical properties of micro-particles obtained during gelation of dispersed phase in phase separated gelatin/pullulan mixtures under well controlled shear conditions.

## 2. Experimental

### 2.1. Preparation of gelled particles

The stock solutions of gelatin and pullulan were prepared by dissolving dry gelatin (UG-719-N, Extraco, Bloom strength 250, Sweden) and pullulan powder (Pullulan 20, molecular weight 200,000, Hayashibara Company Ltd, Japan) in an aqueous 0.1 M NaCl/0.05% sodium azide solution. The dry gelatin powder was first soaked in a jacketed stirred vessel for 30 min at ambient temperature and then dissolved at 60 °C whilst stirring with a helical screw impeller at 250 rpm for 30 min. Pullulan and gelatin mixtures were prepared by mixing the same amount of stock solutions in the jacketed stirred vessel fitted with the helical screw impeller and each mixture was equilibrated at 40 °C for 30 min while been stirred at 250 rpm. Resulting two phase dispersions gelatin rich (G-r) and pullulan rich (P-r) phases were separated at 40 °C by centrifugation at 50,000 g for 2.5 h. The following mixtures were used in this work: 5% gelatin/5% pullulan, 7.5% gelatin/7.5% pullulan and 10% gelatin/10% pullulan giving different composition of gelatin-rich phase as shown in Table 1 (Ding et al., 2005).

The gelatin-rich phase at 1% v/v was dispersed in pullulan rich phase and the dispersion was sheared in a computer controlled Couette device at 1.8 $s^{-1}$ for 60 min at 40 °C. After that time the shear was stopped to avoid aggregation and the temperature was reduced to 20 °C. This led to gelation of the gelatin-rich phase resulting in soft solid particles uniformly suspended in a liquid, pullu-

Table 1
Composition (wt%) and the gelation temperatures of the micro-particles

| | 5%/5% | 7.5%/7.5% | 10%/10% |
|---|---|---|---|
| $X_{\text{gelatin}}$ (%) | 9.5 | 14.7 | 20.0 |
| $X_{\text{pullulan}}$ (%) | 0.48 | 0.34 | 0.16 |
| $T_g$ (°C) | 25.7 | 28.1 | 30.4 |