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Improving encapsulation efficiency and stability of water-in-oil-in-water emulsions using a modified gum arabic (Acacia (sen) SUPER GUM^{TM})

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Abstract

A matured gum arabic, (Acacia (sen) SUPER GUM^{TM}) was investigated as an emulsifier to aid in the stabilisation of water-in-oil-in-water (W/O/W) emulsions. Emulsions were characterised by phase separation, confocal microscopy, droplet sizing, and encapsulation efficiencies (EE). Initial results showed that some precipitates were observed when SUPER GUM^{TM} was dissolved in a 0.1 M sodium phosphate buffer and this necessitated the preparation of emulsions in distilled water systems. Interestingly, the ionic environment had an effect on the emulsifying ability of the synthetic hydrophobic emulsifier, polyglycerol ester of polyricinoleic acid (PGPR). The concentration of PGPR could be reduced to 2 wt% in distilled water systems with an EE > 90%, while in buffered systems, 4 wt% PGPR was required to maintain a similar EE. SUPER GUM^{TM} was added as an emulsifier to replace sodium caseinate either in the internal or external aqueous phase of W/O/W emulsions over a range of concentrations. It was found that SUPER GUM^{TM} destabilised the W/O/W emulsions prepared with low PGPR concentration when added to the internal aqueous phase. On the other hand, addition of 10 wt% SUPER GUM^{TM} to the external aqueous phase allowed further reduction in the PGPR concentration to 0.5 wt% whilst an EE > 90% was maintained. W/O/W emulsions formed with SUPER GUM^{TM} were also found to be stable over a wide pH range, compared to W/O/W emulsions stabilised with sodium caseinate, and thus, may be suitable for applications over a wide range of pH values, as may occur during ingestion or incorporation into different food systems.

Keywords: Multiple emulsions; Double emulsions; PGPR; SUPER GUM^{TM}; Sodium caseinate

1. Introduction

Water-in-oil-in-water (W/O/W) emulsions are systems in which the dispersed phase is itself an emulsion. They have immense potential to be used in the food industry as a means of encapsulation or fat reduction. However, the inherent thermodynamic instability of W/O/W emulsions is a major hurdle in their application in food products (Benichou, Aserin, & Garti, 2004; Hino, Shimabayashi, Tanaka, Nakano, & Okochi, 2001).

A number of factors have been identified as affecting the stability of W/O/W emulsions. These include the method of preparation, the composition of the emulsion, i.e. the nature of oil phase, type of emulsifiers and the nature of entrapped materials as well as the presence of electrolytes. Other parameters such as phase volume, concentration of components also need to be considered (Florence & Whitehill, 1985; Omotosho, Whateley, Law, & Florence, 1986).

Polyglycerol ester of polyricinoleic acid (PGPR) has been regularly used in studies concerned with W/O/W emulsions, as it appears to be the most effective hydrophobic emulsifier available (Wilson, Van Schie, & Howes, 1998). However, the use of this synthetic emulsifier is strictly regulated thus reducing the use or partially substituting PGPR with other food grade materials is desirable. Macromolecular materials (e.g. proteins or polysaccharides), due to their stability at the interface or their functions as stabiliser, can be used in W/O/W emulsion alone or in combination with monomeric materials as emulsifiers in conjunction with PGPR (Evison,

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Dickinson, Owusu, & Williams, 1995). Proteins and polysaccharides may be included in the internal or/and external aqueous phase of W/O/W emulsion to improve the encapsulation efficiency (EE) and stability (Benichou et al., 2004; Garti, Aserin, & Cohen, 1994; Hino et al., 2001).

Studies have shown that BSA improved the stability of W/O/W emulsions when added to the internal aqueous phase to partially replace monomeric emulsifiers (Dickinson, Evison, Owusu, & Zhu, 1993; Evison et al., 1995; Garti et al., 1994). Koberstein-Hajda and Dickinson (1996) incorporated unmodified faba protein into the inner aqueous phase of W/O/W emulsions using Span 80 as hydrophobic emulsifier and sodium caseinate as hydrophilic emulsifier with little change in resulting EE values. Other proteins and polysaccharides used as substitutes of monomeric emulsifiers were gelatine (Vaziri & Warburton, 1995), xanthan gum (Dickinson et al., 1993; Evison et al., 1995) and cyclodextrins (Yu et al., 1999).

Results from our laboratory have shown that sodium caseinate can improve the EE and stability of W/O/W emulsions when incorporated into the internal aqueous phase (Su, Flanagan, Hemar, & Singh, 2006). The addition of 0.5 wt% sodium caseinate to the internal aqueous phase, combined with 2 wt% PGPR, resulted in similar stability and EE compared to emulsions formed with 4 wt% PGPR alone. The results suggested a synergistic effect between the PGPR surfactant in the oil phase and sodium caseinate, which is mainly in the aqueous phase.

Gum arabic is a versatile ingredient used in food systems due to its molecular flexibility (Islam, Phillips, Sljivo, Snowden, & Williams, 1997). Functions of gum arabic include emulsification, encapsulation, stabilisation, water binding, adhesion, film forming and dietary fibre (Dickinson & Galazka, 1991; McNamee, O'Riordan, & O'Sullivan, 1998; Verbeken, Dierckx, & Dewettinck, 2003). However, there have been no reports on the effects of any gum arabic product on W/O/W emulsion systems.

Numerous physicochemical studies of gum arabic have shown that it is heterogeneous, having either a variation in monomer composition or a variation in the mode of linking and branching for the monomer units, in addition to a distribution in molecular mass (Dickinson & Galazka, 1991; Islam et al., 1997; Randall, Phillips, & Williams, 1988; Williams, Phillips, & Stephen, 1990). Gum arabic consists of a major fraction of lower molecular mass (2.5×10^5 Da) which contains very little nitrogeneous material, and arabinogalactan, a high molecular-weight fraction ($1-2.5 \times 10^6$ Da) representing about 10–30% of the total gum and which is associated with a small protein component (Dickinson & Galazka, 1991; Dickinson, Galazka, & Anderson, 1991; Randall et al., 1988).

Amino acid compositions in gum arabic have been studied in detail, as it has been demonstrated that it is the protein containing high molecular-weight fraction which adsorbs most strongly at the oil–water interface (Dickinson & Galazka, 1991; Dickinson et al., 1991; Islam et al., 1997; Street & Anderson, 1983). It appears, however, that the emulsifying behaviour of gum arabic depends not only on the nitrogen content, but also on the molecular accessibility of the protein component for rapid adsorption, and on the distribution of protein between lower and higher molecular-weight fractions (Dickinson, Murray, Stainsby, & Anderson, 1988).

It has been suggested that protein associated high molecular-weight fractions (arabinogalactan) are formed by aggregation of smaller arabinogalactan into larger units. The proportion of large and small arabinogalactan units in gum arabic has been reported to have a direct effect on the emulsifying properties of gum arabic, with a high proportion of larger units desired (Randall et al., 1988; Randall, Phillips, & Williams, 1989; Williams et al., 1990). Dickinson, Elverson, and Murray (1989, 1991) suggested that gum arabic forms a thick, sterically stabilising layer around emulsion droplets, which results in a stable emulsion.

The effect of a modified gum arabic product, referred to as SUPER GUM^{TM}, on the formation and stabilisation of W/O/W emulsion is investigated in the present study. Compared to conventional gum arabic, SUPER GUM^{TM} has undergone an accelerated aggregation process, from which, the smaller arabinogalactan units, join into larger molecular-weight arabinogalactan protein aggregates. While the product is chemically and molecularly identical to the original gum, the proportion of arabinogalactan protein emulsifying component is up to more than double; this results in a significant increase in the interfacial surface properties and stability of the oil droplet in oil-in-water emulsions (Al-Assaf, Phillips, Aoki, & Sasaki, 2007; Aoki, Al-Assaf, Katayama, & Phillips, 2007).

The objective of this study is to investigate the effect of different concentrations of SUPER GUM^{TM} in the internal or external aqueous phases on the formation and stabilisation of W/O/W emulsions, and to determine the effects of an ionic environment on the emulsifying ability of PGPR.

2. Materials and methods

Commercial soybean oil was purchased from AMCO Ltd., Auckland, New Zealand. PGPR (4150, Palsgaard Ltd., Denmark) was purchased from Hawkins Watt Ltd., Auckland, New Zealand. Commercial sodium caseinate was obtained from Fonterra Cooperative Ltd., Palmerston North, New Zealand, and contained approximately 93% protein, 1.2% sodium and 0.06% calcium. Violet dye poly R-478 was obtained from Sigma Ltd., San Louis, USA, and Nile Blue was from BDH Ltd., Poole, UK. SUPER GUM^{TM} was gifted from San-Ei Gen F.F.I. Inc., Tokyo, Japan, and contained approximately 1.7% salt.

2.1. Preparation of W/O/W emulsions

W/O/W emulsions were prepared by a two-step process with a two stage homogeniser (APV, 2000, Denmark) with a slight modification to the method described previously