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ScienceDirect
LWT 40 (2007) 1381–1391
LWT
www.elsevier.com/locate/lwt

Limonene encapsulation in freeze-drying of gum Arabic–sucrose–gelatin systems

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Received 9 April 2006; received in revised form 30 September 2006; accepted 24 October 2006

Abstract

Encapsulation of limonene in freeze-drying of various matrices consisting of gum Arabic, sucrose and gelatin was studied. Retention of limonene in freeze-drying was observed by measuring absorbance at 252 nm using a spectrophotometer. Two different levels of limonene, in the weight ratios (w/w) of 9:1 and 8.5:1.5 (total solids (TS):limonene) were studied. Highest amount of limonene ($75.3 \pm 0.3\%$ of initially added amount) in the emulsions homogenised at 25 MPa (4 MPa in second stage) followed by freeze-drying, was retained in a matrix consisting of gum Arabic, in the ratio of 9:1 (w/w). A mixture consisting of gelatin–sucrose–gum Arabic in the w/w ratio of 0.66:0.17:0.17 retained highest amount ($71.8 \pm 0.1\%$ of initially added amount) of limonene in the ratio (w/w) of 8.5:1.5 (TS:limonene). A matrix consisting of gum Arabic–sucrose–gelatin (1:1:1 w/w/w) with added limonene at a ratio (w/w) of 9:1 (TS:limonene), was used to study the effects of ultra high-pressure homogenisation (50–250 MPa) on limonene encapsulation in freeze-drying. Highest amount (84% of initially added amount) of limonene was retained in the emulsions homogenised at a pressure of 100 MPa. Electron micrograph of freeze-dried matrix of gum Arabic–sucrose–gelatin in the weight ratio of 1:1:1 suggested that it possessed a flake like structure, which was free of dents and shrinkage. A mixture consisting of gum Arabic–sucrose–gelatin is an efficient encapsulant for limonene encapsulation by freeze-drying.

Keywords: Freeze-drying; Encapsulation; Ultra high-pressure (50–250 MPa) homogenisation; Cold stage microscopy

1. Introduction

Encapsulation is typical of dehydration processes, and takes place as a result of formation of an amorphous and continuous matrix around dispersed molecules (Roos, 1995). Encapsulation by entrapment of flavours in a protective continuous matrix allows production of solid matrices with dispersed flavours protected from loss and undesirable changes, such as oxidation (Chang, Scire, & Jacobs, 1988; Rosenberg, Kopelman, & Talmon, 1990; Shahidi & Han, 1993; Sheu & Rosenberg, 1995). Application of encapsulation has become widely accepted in the flavour industry, because most flavours are volatile and chemically unstable in the presence of oxygen, light, water and heat (Bhandari, Dumoulin, Richard, Noleau, & Lebert, 1992; McNamee, O'Riordan, & O'Sullivan, 1998; Risch, 1995).

Different materials, such as carbohydrates and gums, have been used as encapsulation matrices (encapsulants) for flavour encapsulation (Bangs & Reineccius, 1988; Boutboul, Giampaoli, Feigenbaum, & Ducruet, 2002; Levi & Karel, 1995; Reineccius, Reineccius, & Peppard, 2002; Yoshii et al., 2001). An encapsulation matrix should have good solubility in water, emulsifying properties, drying properties and low viscosity at high solids concentration (Kim, Morr, & Schhenz, 1996; Reineccius, 1988). A single encapsulating matrix does not posses all required characteristics and efforts to improve encapsulation properties have been done by using mixtures of carbohydrates with proteins and polysaccharides at different proportions (Bhandari et al., 1992; McNamee, O'Riordan, & O'Sullivan, 2001; Sheu & Rosenberg, 1995; Voilley, 1995).

Gum Arabic is a highly branched polymer with units of galactose, rhamnose, arabinose, and glucuronic acid.

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0023-6438/$30.00
doi:10.1016/j.lwt.2006.10.008

Covalently bound protein fraction within gum Arabic molecules plays a crucial role in its functional properties (Randall, Phillips, & Williams, 1988). Gum Arabic possesses excellent solubility in water and it has surface-active properties, and it produces low-viscosity solutions at high solids concentrations (Thevenet, 1988). Gum Arabic is able to form a dried matrix around dispersed compounds in dehydration processes, which entraps them inside the matrix and prevents volatile loss and contact with air (Thevenet, 1988). These solubility and surface-active qualities have facilitated its extensive use as an encapsulation matrix for retention and protection of chemically reactive and volatile oils and flavours. However, gum Arabic is an expensive ingredient and its availability and costs are subject to fluctuation; hence, there is always a need to evaluate alternative matrices for flavour encapsulation (Gibbs, Kermasha, Alli, & Mulligan, 1999; McNamee et al., 2001).

Investigations to replace gum Arabic with other carbohydrates have been reported (Chattopadhyaya, Singhal, & Kulkarni, 1998; McNamee et al., 2001). Chattopadhyaya et al. (1998) used spray-drying to encapsulate vanillin in mixtures of gum Arabic and oxidised starches. Percentage vanillin encapsulated using oxidised corn starch (60.81%) and an amaranth (58.61%) starch was higher than was encapsulated by gum Arabic (57.41% of initially added amount). It was suggested that oxidised starches in spray-drying could be better encapsulation matrices than gum Arabic. McNamee et al. (2001) studied effects of mixing a range of carbohydrates with gum Arabic on encapsulation of soy oil in spray-drying. It was observed that mixtures of maltodextrin of various dextrose equivalents (DEs) with gum Arabic in ratio of 1:1 (w/w) produced encapsulation levels similar to gum Arabic (74% of initially added amount). However, mixtures of gum Arabic with maize starch resulted in lower levels of retention (30% of initially added amount). McNamee et al. (2001) suggested that maltodextrins could be used in mixtures with gum Arabic for encapsulation of soy oil in spray-drying.

Retention of encapsulated compounds inside various matrices has been investigated in numerous studies. These have addressed factors related to properties of dispersed molecules (Reineccius, 1988; Rulkens & Thijssen, 1967), encapsulation matrices (Inglett, Gelbman, & Reineccius, 1988; McNamee et al., 1998, 2001), and the emulsion (Mongenot, Charrier, & Chalier, 2000; Risch & Reineccius, 1988; Soottitantawat, Yoshii, Furuta, Ohkawara, & Linko, 2003) together with the spray-drying process conditions (Bhandari et al., 1992; Chang et al., 1988; Finney, Buffo, & Reineccius, 2002; Rosenberg et al., 1990; Rulkens & Thijssen, 1967), and the powder morphology during and after drying (Moreau & Rosenberg, 1996) has been reported. Recently, emphasis has been on modifying the properties of emulsion before drying. Droplet size of emulsified material in emulsion is a significant factor for retention of encapsulated compounds. Importance of preparing fine emulsions prior to spray-drying has been investigated in several studies (Mongenot et al., 2000; Risch & Reineccius, 1988; Sheu & Rosenberg, 1995; Soottitantawat et al., 2003). It has been suggested that retention of flavours during spray-drying could be enhanced by reducing the mean emulsion droplet diameter of the dispersed components during emulsification. Risch and Reineccius (1988) studied the effect of emulsion droplet size of orange peel oil emulsified in gum Arabic and 'Amiogum' (modified starch) on retention and shelf-life of orange peel oil. Their results suggested that a small droplet size in emulsion yielded a better retention of the orange oil. Similar results were reported by Sheu and Rosenberg (1995), Mongenot et al. (2000), Soottitantawat et al. (2003).

Different methods can be used commercially to reduce the droplet size of emulsified molecules, namely high speed mixing up to 13,500 RPM, high-pressure (up to 50 MPa) homogenisation, 'microfludisation', and ultrasound emulsification. High-pressure homogenisation is extensively used to emulsify, disperse, mix and in processing of food dispersions. Ultra high-pressure homogenisation up to 350 MPa has received considerable attention since it represents important innovations in texture modifications of emulsions for food products. In comparison to conventional high-pressure (up to 50 MPa) homogenisation, ultra high pressure can also deflocculate clusters of water-insoluble fat fractions and disperse them uniformly. Conventional homogenisation may not be able to achieve energy threshold to break these clusters apart (Floury, Desrumaux, Axelos, & Lardières, 2000). In addition to this, ultra high-pressure homogenisation is also expected to increase the surface activity of emulsifying molecules, thus improving the coating ability of emulsifiers. Emulsifying and rheological properties of emulsions produced by ultra high-pressure homogenisation (up to 350 MPa) have been studied by various researchers (Floury et al., 2000; Floury, Desrumaux, Axelos, & Legrand, 2002; Puppo et al., 2005).

In the present study, we used the mixtures of sucrose and gelatin together with gum Arabic for limonene encapsulation in freeze-drying. Sucrose possesses various properties, such as good solubility in water; non-hygroscopicity, low cost and long shelf-life at ambient temperature and it can be used as a matrix for encapsulation of food ingredients. Sucrose has been used for encapsulation of flavours and oils (Chen, Veiga, & Rizzuto, 1990; Flink & Karel, 1970b). Proteins can stabilise emulsion droplets formed during homogenisation due to their ability to interact with water, small ions and other compounds at oil/water interfaces. Proteins play two major roles: on the one hand, they lower surface tension between the interfaces that are formed during the emulsification process, and on the other hand, they form a macromolecular layer surrounding the dispersed particles which structurally stabilises the emulsions by reducing the rate of coalescence (Walstra, 1988). In food emulsions, stability is usually achieved by the application of proteins as the main stabilisers. Gelatin is a widely used protein for encapsulation (Shahidi & Han, 1993), because of its excellent film-forming properties. It