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Aroma release from gum arabic or egg yolk/xanthan-stabilized oil-in-water emulsions

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ABSTRACT

The partitioning and release of limonene and *trans*-2-hexenal from oil-in-water emulsions was evaluated by applying static headspace gas chromatography. Experiments were carried out on gum arabic/xanthan (GA/X) and egg yolk/xanthan (EY/X)-stabilized emulsions. The dispersed phase (φ_0 = 0.05 and 0.20) was composed of sunflower oil, while the oil droplet size (d_{32}) ranged from 0.8 to 12.5 μm. The type of emulsifying/stabilizing system influenced the thermodynamic component, as expressed by the air–liquid partition coefficient ($K_{a/l}$) values at equilibrium (37 °C), of the release of limonene only. Increase in lipid fraction decreased the release of both aroma compounds. The oil droplet size had no significant impact on the partition coefficients of *trans*-2-hexenal, whereas for a lipid content of 20% (v/v) and GA/X stabilizing system, the partition coefficient of limonene was considerably reduced. Additionally, information on the aroma compound diffusion through the liquid–gas interface was obtained by calculating the initial slopes of the time–release curves. Both aroma compounds diffused more slowly through the GA/X-stabilized emulsions compared to EY/X-stabilized systems, while the release rate of limonene was significantly decreased when oil droplet size was increased. This behaviour was attributed to changes in viscosity as well as to reinforcement of xanthan network because of the presence of gum arabic. On the other hand, the nature of the interfacial film and the extent of oil droplet interactions in the EY/X-stabilized emulsions is considered to be responsible for their lower ability to retain the two aroma compounds. Odour assessment experiments were also conducted and the results were found to partially correlate with gas chromatographic results.

1. Introduction

Aroma release has an important influence on the sensory characteristics and hence consumer's preference of foods. It is generally accepted that aroma molecules stimulate human's chemoreceptor system by reaching the olfactory receptors in the roof of the nasal cavity as well as the gustatory receptors within the oral cavity, via the air and the liquid phases, respectively. Their transfer within the food and their release in the air phase depends on several factors such as their physicochemical properties, their concentration and their interactions with other food constituents. Many studies have been carried out in simple systems composed of water, proteins, lipids and carbohydrates (de Roos, 2000; Fisher & Widder, 1997; Godshall, 1997; Guichard, 2002; Lübbers, Landy, & Voilley, 1998; Roberts, Stephen Elmore, Langley, & Bakker, 1996; Secouard, Malhiac, Grisel, & Decroix, 2003; Terta & Paraskevopoulou, 2006).

However, many food products are two-phase systems, being oil-in-water or water-in-oil emulsions, for instance salad dressings, creams, milk, butter, etc. In emulsions the aroma compounds may be distributed in more than one phase: the aqueous phase, the oil phase and the interface (Druaux & Voilley, 1997). Before they can be released in the air phase, they have to diffuse from one phase to the other (de Roos, 2000). The affinity of the volatiles for these phases as well as the nature and the amount of the dispersed phase (oil or water), the nature and the area of the oil–water interface and the type of emulsifier/stabilizer used are considered to influence the aroma release from emulsified systems (Charles, Rosselin, Beck, Sauvageot, & Guichard, 2000; Doyen, Carey, Linforth, Marin, & Taylor, 2001; Meynier, Lecoq, & Genot, 2005; Miettinen, Tuorila, Piironen, Vehkalahti, & Hyvönen, 2002; Rabe, Krings, & Berger, 2003; Seuvre, Philippe, Rochard, & Voilley, 2006; van Ruth, de Vries, Geary, & Giannouli, 2002a; van Ruth, King, & Giannouli, 2002b).

The role of lipids in flavour perception has already been reported. Their performance as flavour modulators may be attributed to air–liquid partition phenomena as well as to the high resistance to mass transfer, both of them determining the rate at which equilibrium is achieved and the extent of flavour release (Buttery, Guadagni, & Ling, 1973; Druaux & Voilley, 1997). Non-polar aroma compounds are mainly influenced by the presence of lipids and their volatility decreases with increasing lipid level contrary to that

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of polar compounds (Buttery et al., 1973; Guichard, 2002). Variation of the food composition, e.g. fat level reduction, leads to modifications of the structure and the interactions between volatile and non-volatile constituents. Low-fat food formulations are nowadays widely available and their aroma profile is significantly changed compared to that of the respective traditional food products (de Roos, 2000).

Many stabilizers (emulsifiers and/or thickening agents), normally used to physically stabilize emulsion droplets against aggregation, are capable of binding flavour molecules and therefore altering their distribution within an emulsion (Bakker, 1995). Proteins are surface-active molecules which adsorb to the surface of freshly formed droplets, forming a protective membrane, which prevents the droplets from coming close enough together to aggregate, while polysaccharides are ingredients used to increase the viscosity of the continuous phase of emulsions and they enhance emulsion stability by retarding the movement of droplets (McClements, 1999). Interactions of flavour compounds with both types of biopolymers are known to have a strong influence on the release of flavour from foods. Proteins often cause a decrease in the volatility of flavour compounds by interacting with volatiles through reversible hydrophobic bond formation (Lübbers et al., 1998; Rogacheva, Espinosa-Diaz, & Voilley, 1999; Sostmann & Guichard, 1998). Polysaccharides influence the volatility of the aroma molecules and their partitioning between different phases mainly by two mechanisms: The first one is diffusion decrease as predicted by the Stokes–Einstein equation where diffusion is inversely proportional to viscosity (Baines & Morris, 1987). The second mechanism involves specific molecular interactions of the aroma compounds molecules with the macromolecule often due to adsorption, entrapment in microregions, complexation, encapsulation and hydrogen bonding, as in the case of starch (Arvisenet, Le Bail, Voilley, & Cayot, 2002; Conde-Petit, Escher, & Nuessli, 2006; Godshall, 1997).

The study of compositional and structural properties of oil-in-water emulsions on aroma release has led to conflicting conclusions. According to Rabe et al. (2003), the emulsion droplet diameter showed no significant effect on the dynamic flavour release of 13 aroma compounds from different chemical classes. Le Thahn, Thibeaudeau, Thibaut, and Voilley (1992) and Druaux and Voilley (1997) also failed to find an effect, but Charles et al. (2000) and Miettinen et al. (2002) found an influence of the droplet size that depended on the water solubility of the aroma compound. On the other hand, van Ruth et al. (2002a, 2002b) observed that particle size distribution affected the release of almost all studied compounds. According to their observations, the large particle size was related with increased aroma release which affected the thermodynamic as well as the kinetic component of aroma release. The importance of the emulsifier and the emulsion structure on aroma release was also shown by differences in air phase concentrations of aroma compounds in dispersed and non-dispersed biphasic systems. It was reported that the effect of the type of emulsifier on the aroma release was dependent on the specific compound (Landy, Courthaudon, Dubois, & Voilley, 1996).

The investigation of the impact of a food constituent on the retention or release of a volatile compound usually involves the use of a headspace GC technique. Application of static headspace methods has given insight into the partitioning of flavours between the liquid and the air phase. Trapping of headspace volatiles, using porous polymer absorbents, as well as solid phase microextraction have also been used for the analysis of aroma release (Pawliszyn, 1997). Dynamic headspace analysis provides information about the mass transfer behaviour and the temporal release of aroma compounds. However, flavour release in the mouth represents a dynamic situation and the use of MS-nose techniques, simulating mouth conditions, has provided data of high quality (Taylor & Linforth, 2000; Yeretzian, Jordan, Brevard, & Lindinger, 2000).

The aim of the present study was to examine the aroma release of limonene and *trans*-2-hexenal from gum arabic or egg yolk/xanthan oil-in-water emulsions. The two volatiles were chosen because they are important flavour compounds in several lipid-containing foods. Gum arabic as well as egg yolk are two very common emulsifiers used in many food formulations and their impact on aroma release from emulsions stabilized with xanthan gum has not been systematically investigated so far. An effort was made to elucidate the effect of lipid fraction and mean oil droplet diameter of oil-in-water emulsions on aroma release by measuring the air–liquid partition coefficients under equilibrium for the two aroma compounds. Furthermore, a kinetic study of the aroma compound release from the emulsions was carried out to investigate the effect of the emulsion characteristics on aroma compound transfer within the emulsion to the air phase. Finally, the odour assessment of each compound by panellists was assessed and compared with the instrumental results.

2. Materials and methods

2.1. Materials

Gum arabic (GA) and xanthan gum (X) were purchased from Sigma Chemical Co. (USA). Liquid egg yolk (EY) was obtained by first breaking fresh hen's eggs and, following complete removal of the adhering white by rolling the intact yolks on tissue paper, the vitelline membrane was punctured and the liquid yolks were collected, pooled and stored at 4 °C in the presence of 300 mg/L sodium azide (Riedel-de-Haën, Seelze, Germany). Refined sunflower oil was purchased from the local market. Citric acid monohydrate was provided by Merck (Darmstadt, Germany).

The matrices were flavoured with *R*-(+)-limonene [(*R*)-4-isopropenyl-1-methyl-1-cyclohexene; purity ⩾ 96%] and *trans*-2-hexenal (purity ⩾ 97%) (Sigma–Aldrich, Steinheim, Germany), both dissolved in ethanol. The two compounds were chosen because their hydrophobicity is very different to each other. As shown in Table 1, limonene has lower water solubility and is more hydrophobic (higher *n*-octanol/water partition coefficient log*P*) than *trans*-2-hexenal.

2.2. Emulsion preparation

Oil-in-water emulsions, stabilized either by GA (1% w/v)/X (0.3% w/v) or EY (2% w/v)/X (0.3% w/v) mixture, were prepared. Sunflower oil (5 or 20%, v/v) was added in citric acid (10 g/L) aqueous solution of pH 3.8 containing the above-mentioned mixtures under continuous stirring. In order to generate emulsions varying in oil droplet size the crude emulsions were homogenized by employing either in APV (Type: 0401005) pressure homogenizer (Albertslund, Denmark) or in Ultra-Turrax T25 mechanical homogenizer (IKA

Table 1
Physicochemical characteristics[a] of aroma compounds

	Limonene	*trans*-2-Hexenal
Formula	$C_{10}H_{16}$	$C_6H_{10}O$
Molecular weight (g/mol)	136.24	98.15
Boiling point (°)	176.00	146.50
Log*P*	4.57	1.58
Vapour pressure (mmHg) at 25 °C	1.98	6.60
Henry coefficient (atm m^3/mol)	2570×10^{-5}	9.88×10^{-5}
Solubility in water (mg/L) at 25 °C	13.80	5260
Odour description	Citrus	Herbal-green

[a] Syracuse Research Corporation (2005).