



Bubble dynamics in various commercial sparkling bottled waters



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ABSTRACT

Observations were made relevant to common situations involving the service of various sparkling waters. Bubble dynamics and progressive losses of dissolved CO₂ were closely examined in three various batches of carbonated waters holding different levels of CO₂. During the turbulences of the pouring process, a cloud of bubbles appears in the water bulk. Under the action of buoyancy, bubbles progressively reach the free surface, and the cloud of bubbles finally vanishes. Bubbles also nucleate on the glass wall, where they grow by diffusion until buoyancy forces them to detach and rise to the free surface to release their CO₂. The three batches of sparkling waters were clearly differentiated with regard to their bubbles dynamics and losses of dissolved CO₂. Our observations were systematically rationalized and discussed on the basis of mass transfer considerations including molecular diffusion, basic concepts of gas solution thermodynamic equilibrium, and bubble dynamics.

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1. Introduction

In the past 15 years, the global bottled water market has seen a remarkable growth (Euzen, 2006; Storey, 2010; Rani et al., 2012), thus raising in turn legitimate environmental concerns regarding the waste management sector (Gleick, 2010). The Forbes magazine even declared that bottled water is expected to become the largest segment of the U.S. liquid refreshment beverage market by the end of this decade (Forbes, 2014). In 2011, the global bottled water market has reached 233 billion liters sold all over the world (Rodwan, 2012).

Among the global bottled water, the sparkling water segment represents nowadays about 10% of the whole bottled water industry. Nevertheless, this percentage may vary a lot from country to country. In the UK, it is close to the global average, whereas in Germany, which is the biggest bottled water market in the world for premium waters, around 80% of the market is actually sparkling waters (Euzen, 2006). Sparkling waters are often seen as a substitute for sweet beverages, and this is particularly true for flavored sparkling waters (Rani et al., 2012). Suffice to say that the bottled sparkling water is a booming, but very competitive market, involving numerous companies throughout the world, with Europe being the largest producer (75%), followed by the USA (20%) (Bruce, 2013).

Classification and labeling of bottled carbonated waters must be in conformity with EU regulations (E. Directive 2009/54/EC and 2003/40/EC). Commercial bottled carbonated natural mineral waters fall into three categories: (1) "naturally carbonated natural mineral water", when the water content of carbon dioxide coming from the spring, and in the bottle are the same as at source; (2) "natural mineral water fortified with gas from the spring" if the content of carbon dioxide comes from the same resource, but its content in the bottle is greater than the one established at source; and (3) "carbonated natural mineral water" if carbon dioxide from an origin other than the groundwater resource is added. Actually, a method using gas chromatography-isotope ratio mass spectrometry has been proposed to determine the carbon isotope ratio ¹³C/¹²C of CO₂ (Calderone et al., 2007). This method was successfully applied to differentiate whether or not gaseous CO₂ in the headspace of a bottled carbonated water originates from the source spring or is of industrial origin.

The capacity of CO₂ to get dissolved in water is ruled by the well-known Henry's law, which states that the equilibrium concentration *c* of dissolved CO₂ is proportional to the partial pressure of gas phase CO₂ denoted *P*:

$$c = k_H P \quad (1)$$

with *k_H* being the strongly temperature-dependent Henry's law constant of gaseous CO₂ in water (i.e., its solubility) (Carroll and Mather, 1992; Diamond and Akinfief, 2003). Under identical conditions of temperature, water can therefore hold different levels of

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Nomenclature

c_L	concentration of dissolved CO ₂ in the liquid phase, in g L ⁻¹	k_H	Henry's law constant of dissolved CO ₂ in water (i.e., its solubility), in g L ⁻¹ bar ⁻¹
c_0	concentration of dissolved CO ₂ in Henry's equilibrium with gas phase CO ₂ in the bubble, in g L ⁻¹	m	cumulative mass of CO ₂ escaping the liquid phase, in g
c_i	initial concentration of dissolved CO ₂ in the liquid phase, in g L ⁻¹	M	molar mass of CO ₂ , =44 g mol ⁻¹
d	bubble diameter, in m	n	mole number of gaseous CO ₂ in the bubble, in mol
D	diffusion coefficient of dissolved CO ₂ in the liquid phase, in m ² s ⁻¹	P	pressure, in Pa
F_T	total volume flux of gaseous CO ₂ escaping the liquid phase, in cm ³ s ⁻¹	r	bubble radius, in m
g	gravity acceleration, in m s ⁻²	R	ideal gas constant, =8.31 J K ⁻¹ mol ⁻¹
h	level of liquid in the glass, in m	t	time, in s
J	molar flux of gaseous CO ₂ which crosses the bubble interface, in mol ⁻¹ m ⁻² s ⁻¹	T	temperature, in K
k	growth rate of bubbles growing through molecular diffusion in the liquid phase supersaturated with dissolved CO ₂ , in m s ⁻¹	U	ascending bubble velocity, in m s ⁻¹
		v	bubble volume, in m ³
		V	volume of liquid poured into the glass or plastic goblet, in L
		λ	thickness of the diffusion boundary layer around the bubble, in m
		η	dynamic viscosity of water, in Pa s
		ρ	density of water, in kg m ⁻³

dissolved CO₂, depending on the pressure of gas phase CO₂ found in the headspace below the cap or screw cap.

In carbonated beverages, the concentration of dissolved CO₂ is indeed a parameter of paramount importance since it is responsible for the very much sought-after fizzy sensation, and bubble formation (the so-called *effervescence*). In sparkling waters, and carbonated beverages in general, homogeneous bubble nucleation (*ex nihilo*) is thermodynamically forbidden (Wilt, 1986; Lubetkin, 2003). In order to nucleate, bubbles need preexisting gas cavities immersed in the liquid phase, with radii of curvature larger than a critical size. In carbonated beverages typically holding several grams per liter of dissolved CO₂, the critical radius needed to initiate bubble nucleation (under standard conditions for pressure and temperature) is of order of 0.1–0.2 μm (Liger-Belair, 2014). This non-classical heterogeneous bubble nucleation process is referred to as type IV nucleation, following the classification by Jones et al. (1999). The presence of dissolved CO₂ therefore directly impacts consumers of sparkling waters, by impacting several emblematic sensory properties such as (i) the visually appealing frequency of bubble formation (Liger-Belair et al., 2006), (ii) the growth rate of bubbles ascending in the glass (Liger-Belair, 2012), and (iii) the very characteristic tingling sensation in mouth. Carbonation, or the perception of dissolved CO₂, indeed involves a truly very complex multimodal stimulus (Lawless and Heymann, 2010). During carbonated beverage tasting, dissolved CO₂ acts on both trigeminal receptors (Dessirier et al., 2000; Kleeman et al., 2009; Meusel et al., 2010), and gustatory receptors, via the conversion of dissolved CO₂ to carbonic acid (Chandrashekar et al., 2009; Dunkel and Hofmann, 2010), in addition to the tactile stimulation of mechanoreceptors in the oral cavity (through bursting bubbles). More recently, Wise et al. (2013) showed that the carbonation bite was rated equally strong with or without bubbles under normal or higher atmospheric pressure, respectively. However, a consumer preference for carbonated water containing smaller bubbles has been previously reported in a thorough study on the nucleation and growth of CO₂ bubbles following depressurisation of a saturated carbon dioxide/water solution (Barker et al., 2002). Moreover, it was also clearly reported that high levels of inhaled gaseous CO₂ become irritant in the nasal cavity (Cain and Murphy, 1980; Cometto-Muniz et al., 1987).

For all the aforementioned reasons, monitoring accurately the losses of dissolved CO₂ in a glass poured with sparkling water is

of interest for carbonated waters elaborators. In the past 15 years, the physics and chemistry behind effervescence has indeed been widely investigated in champagne and sparkling wines (for a recent and global overview, see Liger-Belair (2012) and references therein). Nevertheless, and to the best of our knowledge, the bubbling process itself and the release of gaseous CO₂ remained poorly explored in sparkling waters, under standard tasting conditions.

The present article reports experimental observations relevant to common situations involving the service of commercial carbonated natural mineral bottled waters. Bubble dynamics and progressive losses of dissolved CO₂ were closely examined in three various batches of naturally carbonated waters holding different levels of CO₂. Our observations were conducted in real consuming conditions, i.e., in a glass and in a plastic goblet. During the pouring process, a cloud of bubbles nucleate and grow in the water bulk. Under the action of buoyancy, bubbles rise toward the free surface, and the cloud of bubbles progressively vanishes. Bubbles also nucleate on the glass wall, where they grow by diffusion until buoyancy forces them to detach and rise toward the free surface. We explored the above questions with dedicated experiments used to quantify the bubble dynamics, and the kinetics of gaseous CO₂ discharging from the liquid phase (in real consuming conditions) as described in Section 2. In Section 3.1., the lifetime of the quickly vanishing cloud of bubbles following the pouring step is examined. In Section 3.2., the progressive losses of dissolved CO₂ escaping from the liquid phase (once it is poured in a plastic goblet) are measured and discussed. Finally, in Section 3.3., kinetics of bubbles growing stuck on the plastic goblet are closely examined. Our observations are rationalized and discussed on the basis of mass transfer considerations including molecular diffusion, basic concepts of gas solution thermodynamics, and ascending bubble dynamics.

2. Materials and methods

2.1. The three batches of carbonated waters

Three batches of various commercial carbonated natural mineral bottled waters from Poland, and provided by Danone Research, were investigated. They are described and referenced as follows:

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