Kinetics of CO$_2$ Fluxes Outgassing from Champagne Glasses in Tasting Conditions: The Role of Temperature

GÉRARD LIGER-BELAIR,* SANDRA VILLAUME, CLARA CILINDRE, AND
PHILIPPE JEANDET

Laboratoire d'Œnologie et Chimie Appliquée, UPRES EA 2069, URVVC, Faculté des Sciences,
Université de Reims, B.P. 1039, 51687 Reims Cedex 2, France

Measurements of CO$_2$ fluxes outgassing from a flute poured with a standard Champagne wine initially holding about 11 g L^{-1} of dissolved CO$_2$ were presented, in tasting conditions, all along the first 10 min following the pouring process. Experiments were performed at three sets of temperature, namely, 4 °C, 12 °C, and 20 °C, respectively. It was demonstrated that the lower the champagne temperature, the lower CO$_2$ volume fluxes outgassing from the flute. Therefore, the lower the champagne temperature, the lower its progressive loss of dissolved CO$_2$ concentration with time, which constitutes the first analytical proof that low champagne temperatures prolong the drink's chill and helps retains its effervescence. A correlation was also proposed between CO$_2$ volume fluxes outgassing from the flute poured with champagne and its continuously decreasing dissolved CO$_2$ concentration. Finally, the contribution of effervescence to the global kinetics of CO$_2$ release was discussed and modeled by the use of results developed over recent years. The temperature dependence of the champagne viscosity was found to play a major role in the kinetics of CO$_2$ outgassing from a flute. On the basis of this bubbling model, the theoretical influence of champagne temperature on CO$_2$ volume fluxes outgassing from a flute was discussed and found to be in quite good accordance with our experimental results.

KEYWORDS: Champagne; sparkling wines; carbonated beverages; CO$_2$; effervescence; bubble nucleation; champagne tasting

INTRODUCTION

From a strictly chemical point of view, champagne and sparkling wines are multicomponent hydroalcoholic systems supersaturated with CO$_2$-dissolved gas molecules formed together with ethanol during the second fermentation process. Actually, during this second fermentation process, which occurs in cool cellars, the bottles are sealed so that the CO$_2$ molecules cannot escape and progressively dissolve into the wine. Therefore, dissolved CO$_2$ molecules in the wine and gaseous CO$_2$ molecules under the cork progressively establish equilibrium (an application of Henry's law, which states that the partial pressure of a given gas above a solution is proportional to the concentration of the gas dissolved into the solution). Champagne or sparkling wines elaborated according to the *méthode traditionnelle* typically hold about 10 to 12 g/L of dissolved CO$_2$ molecules (*1*). As soon as a bottle of champagne or sparkling wine is uncorked, the liquid instantaneously becomes supersaturated with dissolved CO$_2$ molecules (since ambient air contains only traces of gaseous CO$_2$). To reach a new stable thermodynamic state with regard to CO$_2$ molecules, champagne

must therefore progressively degas. The progressive release of CO$_2$-dissolved gas molecules from the liquid medium is responsible for bubble formation (the so-called effervescence process). It is worth noting that approximately 5 L of gaseous dissolved CO$_2$ must escape from a typical 0.75 L champagne bottle. No wonder champagne and sparkling wine tasting mainly differs from still noneffervescent wine tasting due to the presence of carbon dioxide bubbles continuously rising through the liquid medium. This is the reason why considerable efforts have been conducted during the past few years in order to better illustrate, detect, understand, and finally control each and every parameter involved in the bubbling process (for a review, see, for example, ref *1* and references therein).

Generally speaking, effervescence in a glass of champagne or sparkling wine may have two distinct origins. It can be natural or artificial. On the one hand, natural effervescence is related to the bubbling process from a glass that has not experienced any specific surface treatment. Closer inspection of such glasses with champagne and sparkling wines revealed that most of the bubble nucleation sites were found to be located on pre-existing gas cavities trapped inside hollow and roughly cylindrical cellulose-fiber-made structures on the order of 100 μm long with

* Corresponding author. Tel/Fax: + 333 26 91 86 14. E-mail: gerard.liger-belair@univ-reims.fr.

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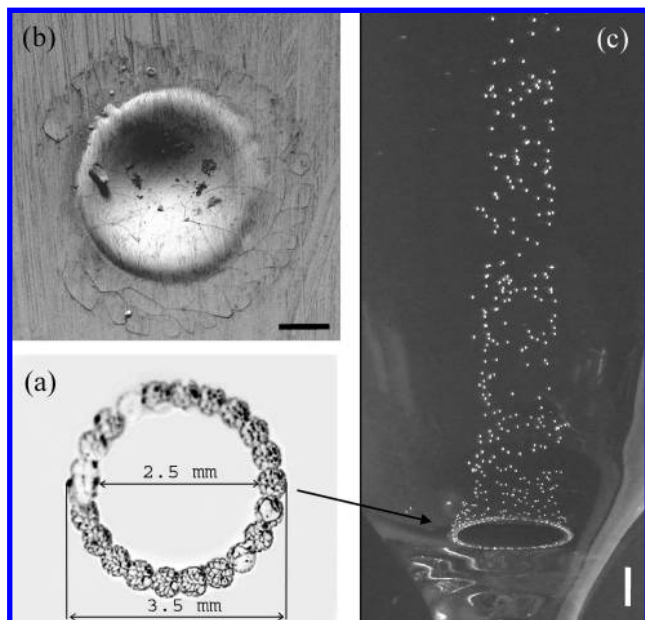
Figure 1. At the bottom of this flute, on its axis of symmetry, the glassmaker has engraved a small ring (done with adjoining laser beam impacts) (**a**). Single laser beam impact as viewed through a scanning electron microscope (bar = 100 μm) (**b**). Effervescence in this flute is promoted from these artificial micro scratches into the form of a characteristic and easily recognizable vertical bubble column rising on its axis of symmetry (**c**) (bar = 1 mm). Reprinted with permission from ref *10*. Copyright 2008 The Royal Society of Chemistry.

a cavity mouth of several micrometers (*2–4*). Natural effervescence may also arise from calcium carbonate crystals precipitated on the glass wall and resulting from the evaporation process after rinsing the glass with tap water. Actually, in a flute showing natural effervescence, bubbles arise from nucleation sites located randomly on the flute's wall. Therefore, there is a substantial variation concerning the natural effervescence between flutes depending on how the flute was cleaned and how and where it was left before serving. On the other hand, artificial effervescence in a glass is related to bubbles arising from scratches intentionally created by the glassmaker to promote effervescence or eventually replace a deficit of natural effervescence (*4–7*). Those microscratches are geometrically able to trap tiny air pockets when champagne is poured into the glass (as cellulose fibers do). Glasses engraved at their bottom are nevertheless indeed easily recognizable, with a characteristic bubble column rising on their axis of symmetry.

From the consumer's point of view, the role of bubbling is indeed essential in champagne, in sparkling wines and even in any other carbonated beverage. Without bubbles, champagne would be unrecognizable, and beers and sodas would be flat. However, the role of effervescence is suspected to go far beyond the solely aesthetical point of view. Actually, in enology, effervescence is believed to play a major role concerning flavor release and CO_2 discharge in glasses containing champagne and sparkling wines. It was demonstrated recently that the continuous flow of ascending bubbles through the liquid medium strongly modifies the mixing and convection conditions of the liquid medium (*7, 8*). In turn, the CO_2 discharge from the liquid surface may be considerably accelerated, as well as the release of the numerous volatile (and potentially aromatic) organic compounds, which strongly depends on the mixing flow conditions of the liquid medium (*7–9*). Glass shape is also suspected to play an important role concerning the kinetics of CO_2 and flavor release during champagne tasting (*7, 8*). From the consumer's point of

view, the release of a sudden and abundant quantity of CO_2 above the champagne surface is known to strongly irritate the nose during the evaluation of aromas. This is the reason why, in recent years, much interest has been devoted to better understand and depict each and every parameter involved in the release of gaseous CO_2 from glasses filled with champagne or sparkling wine. Quite recently, glassmakers showed interest in proposing in the near future to consumers a new generation of champagne tasting glasses, especially designed, with a well-controlled CO_2 release all along tasting (*10*). Accurate and continuous measurement of gaseous CO_2 fluxes discharging from a glass filled with champagne (in tasting conditions) is therefore considered as a first step, helping us to better understand the global kinetics of CO_2 release from glasses filled with champagne or sparkling wine.

In this article, and for the very first time, measurements of CO_2 fluxes outgassing from a flute filled with champagne are presented, in tasting conditions, all along the first 10 min following the pouring process. Since the wine temperature is suspected to play a major role in the kinetics of flavor and gas release in champagne and sparkling wine tasting, experiments were performed at three sets of temperatures, namely, 4 °C, 12 °C, and 20 °C. The contribution of effervescence (i.e., CO_2 bubbles nucleating, ascending, and finally collapsing at the champagne surface) to the global kinetics of CO_2 release was discussed and modeled by the use of results developed over recent years.

MATERIALS AND METHODS

Some Physicochemical Parameters of Champagne. A standard commercial champagne wine was used for this set of experiments, namely a young one, recently elaborated (vintage 2006), and stored in a cool cellar since it was elaborated. Some physicochemical parameters of champagne samples were already determined at 20 °C, with samples of champagne first degassed (*4*). The static surface tension of champagne γ was found to be of the order of 50 mN m^{-1}, its density ρ was measured and found to be close to 10^3 kg m^{-3}, and its dynamic viscosity η was found to be of the order of 1.5×10^{-3} kg m^{-1} s^{-1}. In the range of temperatures that concerns this set of experiments (from 4 to 20 °C), the surface tension and density of champagne do not drastically vary. However, in this range of temperatures, the champagne viscosity η is highly temperature-dependent. In a previous work, the temperature dependence of the champagne viscosity was measured with a thermostatted Ubelhode capillary meter (*11*). In this range of temperatures, the temperature dependence of the champagne dynamic viscosity (expressed in kg m^{-1} s^{-1}) accurately follows an Arrhenius-like equation as follows:

$$\eta(T) \approx 1.08 \times 10^{-7} \exp(2806/T) \qquad (1)$$

with T being the absolute temperature (in K).

Measuring the Initial Concentration of Dissolved CO_2 in the Bottle. The initial concentration of CO_2 molecules dissolved in champagne (before pouring), was determined using carbonic anhydrase. This is the official method recommended by the OIV (namely, the International Office of Vine and Wine) for measuring the CO_2 content in champagne and sparkling wines (*12*). Before opening the bottle, a label was placed at the level of filling of the bottle (in order to measure its volume V precisely). The temperature of the bottle was then decreased until slightly frozen and allowed to warm up again until the ice crystals disappeared. The bottle was opened, and a 50 mL volume of wine (volume v) was rapidly removed and treated apart as a control. Ten milliliters of 12.5 N sodium hydroxide was then directly added to the wine into the bottle. Ten milliliters of this solution (wine + NaOH) was then placed into a 100 mL Erlen-meyer flask containing 30 mL of boiled distilled water together with carbonic anhydrase. Sulfuric acid (0.05 mol L^{-1}) was then added, under agitation, until the pH of the solution reaches first 8.6 and then 4.0 (*n* corresponds to the H$_2$SO$_4$ added volume allowing the pH to decrease from 8.6 to 4.0). The control

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was treated as follows: after CO_2 was removed under vacuum, 10 mL of wine was introduced into a 100 mL Erlen-meyer flask containing 30 mL of boiled distilled water. The pH of this solution was adjusted to 10 with 12.5 N NaOH. A volume n' of 0.05 mol L^{-1} H_2SO_4 was finally added to decrease the pH from 8.6 to 4.0 (see above). This volume measures the acidity of the wine organic acids. The concentration of CO_2 (in g L^{-1}) is then expressed as follows:

$$[CO_2] = 0.44(n - n')\left(\frac{V - v + 20}{V - v}\right) \quad (2)$$

with n, n', V, and v, all being expressed in milliliters. To enable a statistical treatment, six successive CO_2-dissolved measurements were systematically done on six distinct bottles. The champagne elaborated in 2006 was found to initially hold (before pouring) a concentration of CO_2-dissolved molecules of $[CO_2] = 11.3 \pm 0.3$ g L^{-1}.

Glass Washing Protocol. In order to avoid the randomly located bubbling environment inevitably provided in glasses showing natural effervescence, we decided to use, for this set of experiments, a single standard flute engraved on the bottom (thus providing a standardized and artificial effervescence). A rendering of the ring-shaped engravement releasing bubbles at the bottom of the flute is displayed in **Figure 1**. Between the successive pouring and time series data recordings, the flute was systematically thoroughly washed in a dilute aqueous formic acid solution, rinsed using distilled water, and then compressed air-dried. This drastic treatment forbids the formation of calcium carbonate crystals on the flute wall as well as the adsorption of any dust particle acting as natural bubble nucleation sites. Therefore, in this case, the CO_2 bubble nucleation is restricted to the bubble nucleation sites of the ring-shaped engraving so that differences in the kinetics of CO_2 release from one experiment to another are attributed only to physicochemical differences between the champagne samples themselves (namely, differences between the champagne samples' temperature in the present case).

Experimental Setup and Procedure Used to Measure the Flux of CO_2 Desorbing from the Flute Filled with Champagne. A volume of 100 mL of champagne was carefully poured into the flute engraved on its bottom (with an accuracy of ± 4 mL). Immediately after pouring, the flute was then manually placed on the weighing chamber base plate of a precision weighing balance (Sartorius-Extend Series ED, Germany) with a total capacity of 220 g and a standard deviation of ± 0.001 g. The Sartorius balance was interfaced with a laptop PC recording data every 5 s from the starting signal, activated just before the flute filled with champagne was placed on the weighing chamber base plate. The total cumulative mass loss experienced by the flute filled with champagne was recorded during the first 10 min following the pouring process. Actually, the mass loss of the flute poured with champagne is the combination of both (i) champagne evaporation, and (ii) CO_2 progressively desorbing from the supersaturated liquid. The mass loss attributed to champagne evaporation only was accessible by recording the mass loss of a flute filled with a sample of 100 mL of champagne first degassed under vacuum. Because of likely variations in hygrometric conditions from one day to another (occurring even in a climatized temperature-controlled room), champagne evaporation was thus measured with a sample of champagne first degassed under vacuum, just before each series of total mass loss recordings were done. Finally, the cumulative mass loss versus time attributed only to CO_2 molecules progressively desorbing from champagne may therefore easily be accessible by subtracting the data series attributed to evaporation only from the total mass loss data series, as displayed in **Figure 2**, for example.

From a cumulative mass loss−time curve, such as that displayed in **Figure 2**, the mass flux of CO_2 desorbing from the champagne surface (denoted F_{CO_2}) is therefore deduced all along the degassing process in the flute, by dividing the mass loss Δm between two data recordings by the time interval Δt between two data recordings (i.e., $F_{CO_2} = \Delta m/\Delta t$, Δt being equal to 5 s). In other words, the CO_2 mass flux (in g/s) desorbing out of the champagne surface is determined by the slope of the short-dash curve drawn by the cumulative CO_2 mass loss data recordings displayed in **Figure 2**.

In champagne and sparkling wine tasting, it is certainly more pertinent to deal with volume fluxes rather than with mass fluxes of
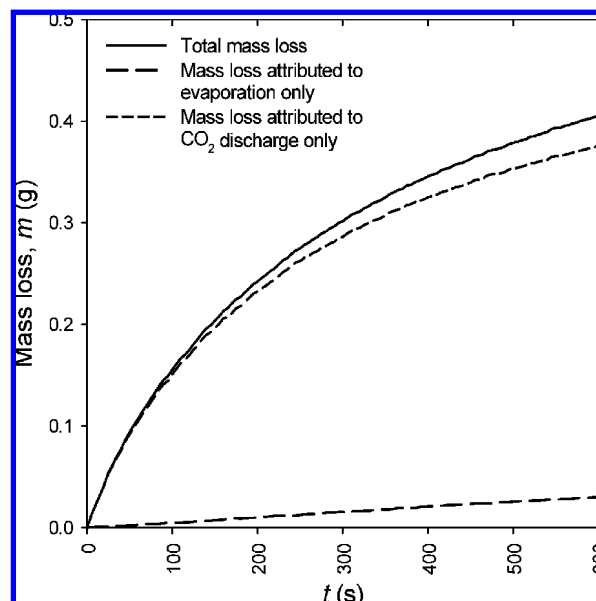
Figure 2. Typical cumulative mass loss recordings (in gram) from a 100 mL flute poured with champagne, all along the first 10 min following the pouring process (at 20 °C). The mass loss data series attributed to CO_2 desorbing only is deduced by subtracting the data series attributed to evaporation only from the total mass loss data series. Consequently, the CO_2 mass flux desorbing from the champagne surface is the slope of the short-dash curve (i.e., the first derivative of the cumulative mass loss data series attributed to CO_2 desorbing only).

CO_2. By considering the gaseous CO_2 desorbing out of the champagne as an ideal gas, the experimental volume flux of CO_2 (in m^3 s^{-1}), denoted F_{exp}, is therefore deduced as follows, all along the degassing process:

$$F_{exp} = \left(\frac{RT}{MP}\right)\frac{\Delta m}{\Delta t} \quad (3)$$

with R being the ideal gas constant (equal to 8.31 J K^{-1} mol^{-1}), T being the champagne temperature (expressed in K), M being the molar mass of CO_2 (equal to 44 g mol^{-1}), P being the ambient pressure (close to 10^5 N m^{-2}), Δm being the loss of mass between two successive data recordings expressed in g, and Δt being the time interval between two data recordings (i.e., 5 s in the present case).

In order to test the influence of champagne temperature on the kinetics of CO_2 release from the flute, experiments were performed at three sets of champagne temperature: 4 °C, 12 °C, and 20 °C (in a temperature-controlled room). To enable a statistical treatment, six successive pourings and time series data recordings were done for each set of temperature (namely, 4 °C, 12 °C, and 20 °C). At each step of the time series (i.e., every 5 s), an arithmetic average of the six data provided by the six successive time series corresponding to a single champagne temperature was done, to finally produce one single average time series, which is characteristic of a given champagne temperature (but with standard deviations corresponding to the root-mean-square deviations of the values provided by the six successive data recordings).

RESULTS AND DISCUSSION

Initial Concentration of Dissolved CO_2 in the Flute, after Pouring. Initially (before pouring), the bulk concentration c_L of dissolved CO_2 was measured by the use of carbonic anhydrase, just after opening the bottle. Nevertheless, the pouring process is far from being consequence-less with regard to c_L. During the several seconds of the pouring process preceding the beginning of the cumulative mass loss−time series, champagne undergoes highly turbulent and swirling flows (*13*). During this phase, champagne looses a very significant

part of its initial content in dissolved CO_2. Consequently, at the beginning of the time series (i.e., at $t = 0$, after the flute was filled with champagne and manually placed on the base plate of the precision weighing balance), CO_2 dissolved in champagne is well below 11.3 ± 0.3 g L^{-1} (as chemically measured inside a bottle, after uncorking, but before pouring). It is simply impossible to analytically determine the CO_2 concentration of champagne inside the flute, after pouring (with the chemical method previously developed in the Materials and Methods section). Because CO_2 molecules continuously desorb from the flute filled with champagne, champagne would have lost a significant quantity of CO_2 before being frozen (as it is a necessary step for measuring dissolved CO_2). Nevertheless, the initial concentration (at $t = 0$) of dissolved CO_2, after the champagne was poured into the flute, may be indirectly accessed by measuring the total cumulative mass loss attributed to CO_2 molecules progressively desorbing from champagne, all along the gas discharging process. The method is similar to that developed earlier, but with time series of the order of 10 h rather than 10 min only so that the cumulative mass loss attributed to CO_2 only finally reaches a strictly horizontal plateau, which betrays the fact that CO_2 desorption completely vanishes through the lack of dissolved-CO_2 inside the liquid matrix. Three successive pouring and cumulative mass loss−time series (during 10 h, up to final degassing) were performed at 20 °C, in order to evaluate the initial concentration of dissolved CO_2, denoted c_i, at the beginning of the time series (at $t = 0$). The total mass of CO_2, denoted m_T, outgassed from the champagne flute was found to be $m_T \approx 0.62 \pm 0.04$ g. In our experiments, champagne was therefore found to hold $c_i = m_T/V_{flute} \approx 6.2 \pm 0.4$ g L^{-1} (i.e., approximately 5 g L^{-1} less than inside the bottle, before pouring). In the preceding relationship, V_{flute} is the volume of champagne poured into the flute expressed in L (namely, 0.1 L in the present case). Therefore, it seems that turbulences of the pouring process cause very significant and quite unexpected loss of dissolved CO_2 during champagne serving.

Influence of Champagne Temperature on Its Loss of Dissolved CO_2 Concentration with Time. The three "average" cumulative mass loss−time series corresponding to three different champagne temperatures are displayed in **Figure 3**, all along the first 10 min following the pouring of champagne into the flute. Despite significant standard deviations (mainly attributed to the difficult repeatability of the manual pouring process between the six successive pouring and time series data records conducted for each temperature), significant differences appear between the three cumulative mass loss−time curves. It is clear from **Figure 3** that the higher the temperature of champagne, the higher the cumulative mass loss of CO_2 with time. In enology, the parameter that characterizes a wine sample with regard to its CO_2 content is its CO_2 concentration, denoted c_L, usually expressed in g L^{-1}. The progressive loss of CO_2 concentration after champagne was poured into the flute, expressed in g L^{-1} and denoted $\Delta c(t)$, may finally easily be accessed by retrieving the mass loss−time curves by using the following relationship:

$$\Delta c(t) = c_L(t) - c_i = -\frac{m(t)}{V_{flute}} \qquad (4)$$

with c_i being the initial concentration of CO_2 dissolved into the champagne after pouring (at $t = 0$), $m(t)$ being the cumulative mass loss of CO_2 with time expressed in g, and V_{flute} being the volume of champagne poured into the flute expressed in L (namely, 0.1 L in the present case).

For each investigated champagne temperature, the corresponding loss of dissolved CO_2 concentration with time along
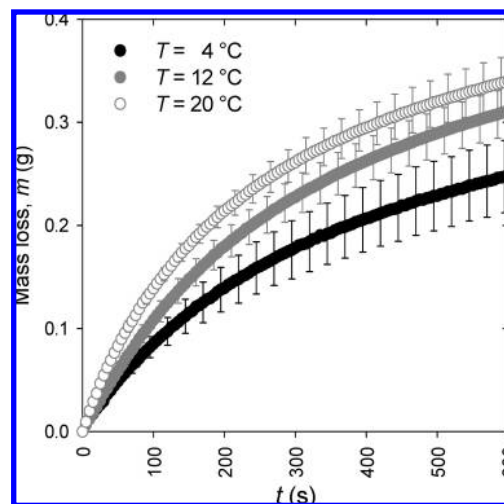
Figure 3. Three average cumulative mass loss-time series corresponding to three different champagne temperatures, all along the first 10 min following the pouring of champagne into the flute. Each data of each cumulative mass loss-time series is the arithmetic average of six successive data issued from six successive pourings. Standard deviations correspond to the root-mean-square deviations of the values provided by the six successive data recordings.
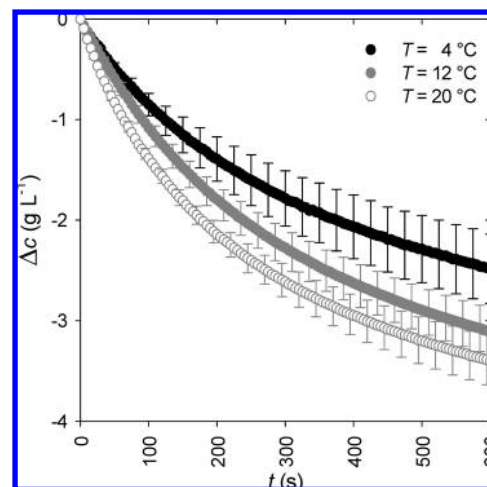
Figure 4. Progressive loss of CO_2-dissolved concentrations with time from a 100 mL flute filled with champagne (in g L^{-1}), as determined with eq 4 all along the first 10 min following the pouring process, and at three different champagne temperatures.

the first 10 min following pouring is displayed in **Figure 4**. It is clear from **Figure 4** that the lower the champagne temperature, the lower its progressive loss of dissolved CO_2 concentration with time. From the taster's point of view, this observation is of importance for both the visual aspect of champagne and its mouth feel sensation. Actually, it was recently shown that the higher the concentration of dissolved CO_2 in champagne, the higher the kinetics of bubble formation, the larger the average bubbles' size, and finally the more effervescence in the flute (*4*). Moreover, it is also well known in champagne and sparkling wine tasting that the higher the concentration of dissolved CO_2, the higher the fizzy sensation when bubbles burst over the tongue (*14*). To the best of our knowledge, this is the first set of analytical results concerning the influence of champagne temperature on its progressive loss of dissolved CO_2 concentrations with time in tasting conditions and therefore the first analytical proof that low champagne temperatures prolong the drink's chill and helps retains its effervescence.

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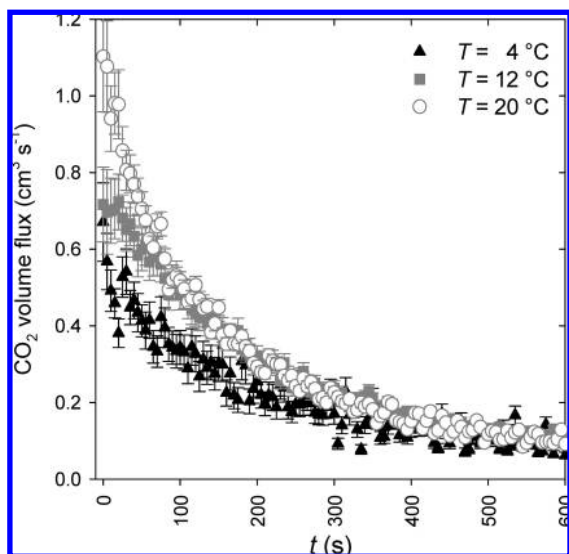
Figure 5. CO_2 volume flux recordings (in cm³ s⁻¹) desorbing from a 100 mL flute filled with champagne, as determined with eq 3, all along the first 10 min following the pouring process, and at three different champagne temperatures. Each data of each time series is the arithmetic average of six successive values recorded from six successive pourings. Standard deviations correspond to the root-mean-square deviations of the values provided by the six successive data recordings.

Influence of Champagne Temperature on CO_2 Volume Fluxes Outgassing from the Flute. In champagne and sparkling wine tasting, in addition to the visual aspect of effervescence and mouth feel, both depending (among many other parameters) on the dissolved-CO_2 concentration $c(t)$, another important aspect is the smell or nose of the wine, the so-called bouquet (*14−16*). Effervescence of champagne and sparkling wines promotes the development of aromas in the headspace above the glass. The myriad bubbles nucleating on the flute's wall and traveling through the wine's bulk considerably enhance the perception of volatile organic compounds by considerably enhancing exchange surfaces between the wine and the atmosphere. It is worth noting that, in a typical flute filled with champagne or sparkling wine, approximately 50,000 bubbles would have already nucleated, risen, and finally exploded during the first three minutes following pouring (*10*). Each bubble having a diameter close to 1 mm, this cloud of 50,000 bubbles in total creates a global exchange surface of approximately 1500 cm² during the first 3 min following pouring. This is huge compared with the area of the air/wine interface, which rarely exceeds 20 cm² in a classical flute. However, each bubble collapsing at the wine's surface inevitably frees its tiny CO_2 volume. Consequently, the inevitable counter party of the exhausting aromas effect attributed to the exchange surfaces of the bubbles is to progressively increase the concentration of gaseous CO_2 in the headspace above the wine's surface. It is indeed well known that a sudden and abundant quantity of CO_2 may irritate the nose during the evaluation of aromas (*14*). The pertinent analytical parameter, which characterizes the progressive release of gaseous CO_2 desorbed from a flute filled with champagne, is the volume flux of CO_2 escaping from the wine/ air interface, as defined in eq 3. The CO_2 volume fluxes outgassing from the flute filled with champagne, as determined with eq 3 all along the first 10 min following pouring and for each champagne temperature, are presented in the graph displayed in **Figure 5**. Experimentally, it is clear that the lower the champagne temperature, the lower CO_2 volume fluxes
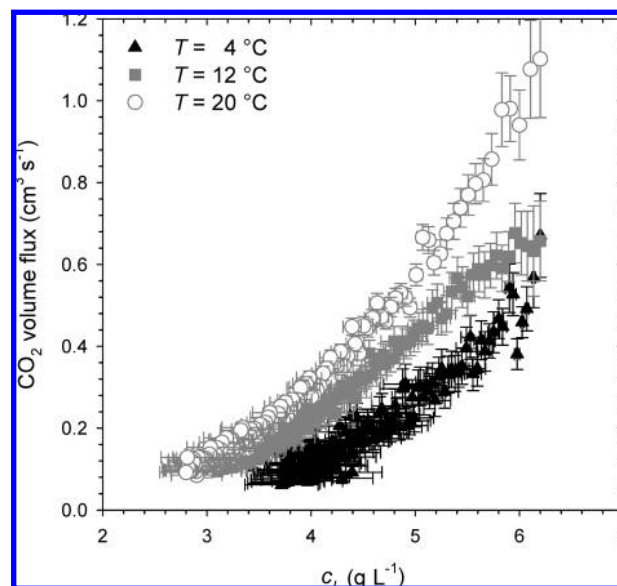
Figure 6. CO_2 volume flux recordings (in cm³ s⁻¹) desorbing from a 100 mL flute filled with champagne as a function of its dissolved-CO_2 concentration, and at three different champagne temperatures. Each data of each time series is the arithmetic average of six successive values recorded from six successive pourings.

outgassing from the flute and especially in the early moments following the pouring process.

Generally speaking, the driving force behind the desorption of gas species dissolved into a supersaturated liquid medium is the difference between the bulk concentration c_L of dissolved gas molecules and the Henry's law equilibrium concentration c_{eq}. In the case of champagne, after opening the bottle, the equilibrium concentration c_{eq} of CO_2-dissolved gas molecules must fulfill the following Henry's law expressed hereafter:

$$c_{eq} = k_H P_{CO_2} \qquad (5)$$

with P_{CO_2} being the partial pressure of CO_2 molecules in ambient air, and k_H being the Henry's law constant (i.e., the solubility of CO_2 molecules in the liquid medium). The partial pressure of CO_2 above champagne being in the order of only 0.0004 bar (since the natural abundance of CO_2 in the ambient air is close to 400 ppm), and the solubility of CO_2 in champagne being of the order of 1.5 g/L/bar, at 20 °C (*1*), the equilibrium CO_2 concentration can be expressed as $c_{eq} = 1.5 \times 4 \times 10^{-4} \approx 0.6$ mg L⁻¹ $\ll c_L$. Therefore, the difference Δc between the bulk concentration c_L and the equilibrium concentration c_{eq} of dissolved gas molecules becomes $\Delta c = c_L - c_{eq} \approx c_L$. Finally, once poured into the flute, the driving force behind the desorption of CO_2 from champagne being its bulk concentration c_L of dissolved CO_2, it seemed pertinent to propose a correlation between the CO_2 volume flux outgassing from the flute and the continuously decreasing bulk concentration c_L of dissolved CO_2. To do so, time series data recordings displayed in **Figures 4** and **5** were combined. Time was eliminated so that the CO_2 volume flux outgassing from the flute was plotted as a function of champagne dissolved CO_2 concentration c_L. Correlations between CO_2 volume fluxes outgassing from the flute and dissolved CO_2 concentrations in champagne are displayed in **Figure 6**, for each champagne temperature. It is clear from **Figure 6** that, for a given dissolved CO_2 concentration of champagne, the lower the champagne temperature, the lower CO_2 volume fluxes outgassing from the flute.

In the following paragraph, a multiparameter bubbling model was built upon results obtained over recent years. The theoretical

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influence of champagne temperature on CO_2 volume fluxes released into the form of CO_2 bubbles rising and collapsing at the liquid surface is discussed.

Modeling CO_2 Volume Fluxes Outgassing from the Flute: The Contribution of Bubbles and the Role of Temperature. Actually, the contribution of rising CO_2 bubbles to the global kinetics of CO_2 outgassing from a sparkling beverage is quite easily accessible by taking into account the whole number of nucleation sites found in the glass, the average frequency of bubble production from a nucleation site, and the average size of a bubble as it reaches the liquid surface (to finally collapse and release its CO_2 content in the headspace above the liquid surface). Recently, models based on both classical diffusion and ascending bubble velocity were developed in order to propose scale laws likely to link the frequency of bubble nucleation (i.e., the number of bubbles released per second from a given nucleation site) as well as the size of CO_2 bubbles rising in a carbonated beverage with some physicochemical parameters of the liquid medium. The frequency of bubble formation from a single bubble nucleation site, denoted f, was found to obey the following scaling law (*11*):

$$f \propto \frac{T^2(c_L - k_H P)}{\eta P} \tag{6}$$

with T being the liquid temperature (in K), c_L being the bulk concentration of CO_2 in the liquid medium, k_H being the so-called Henry's law constant (i.e, the solubility of the CO_2 molecules with regard to the liquid medium), P being the ambient pressure, and η being the liquid dynamic viscosity.

The diameter of a bubble reaching the liquid surface was also found to depend on various parameters (*1, 4*). Actually, the diameter of a bubble, denoted d, was found to obey the following scaling law:

$$d \propto T^{5/9}\left(\frac{1}{\rho g}\right)^{2/9}\left(\frac{c_L - k_H P}{P}\right)^{1/3} h^{1/3} \tag{7}$$

with ρ being the liquid density, g being the acceleration due to gravity, and h being the distance traveled by the bubble from its nucleation site. The growth of bubbles has also been dealt with recently in case of ascending champagne and beer bubbles (*17*), using the more general theoretical development of Zhang and Xu (*18*). More on convective bubble and on crystal growth and dissolution can be found in a recent textbook by Professor Zhang (*19*).

Finally, the contribution of effervescence to the global CO_2 volume fluxes outgassing from a sparkling beverage poured in the engraved flute may be accessed by multiplying the number N of nucleation sites found in the flute by the average frequency f of bubble nucleation and by the average volume v of a bubble collapsing at the liquid surface. Therefore, by combining the two above-mentioned scaling laws, the CO_2 volume flux released by bubbles rising and collapsing in a carbonated beverage poured into a glass, denoted dV/dt, obeys the following scaling law:

$$\frac{dV}{dt} = Nfv \propto Nfd^3 \propto N\frac{T^{11/3}}{\eta}\left(\frac{1}{\rho g}\right)^{2/3}\left(\frac{c_L - k_H P}{P}\right)^2 h \tag{8}$$

It is worth noting from the previous relationship that, every other parameter being equal, the higher the bulk concentration c_L of CO_2 in the liquid medium, the higher the CO_2 volume flux outgassing from the liquid into the form of CO_2 bubbles collapsing at the liquid surface (as experimentally shown in **Figure 6**).
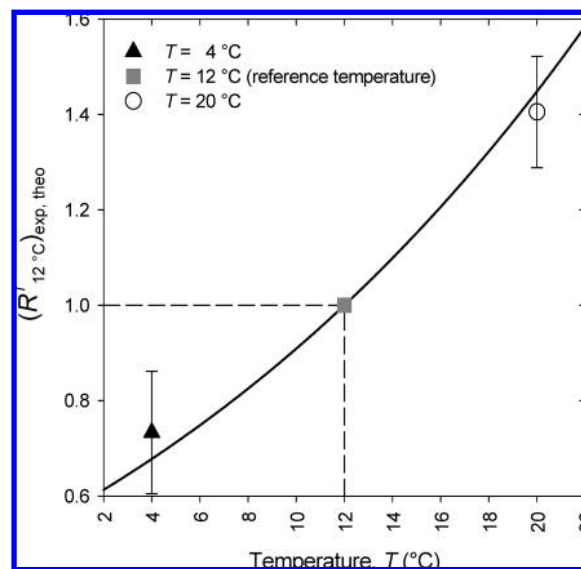

Figure 7. Experimentally determined ratios, $(R_{12^\circ C}^{4^\circ C})_{exp}$ and $(R_{12^\circ C}^{20^\circ C})_{exp}$ (obtained just after pouring, i.e., at $t = 0$), which both compare the average CO_2 volume fluxes outgassing from champagne, at 4 and 20 °C, respectively, with the average CO_2 volume flux outgassing from champagne at the reference temperature of 12 °C. In order to test the temperature dependence of our outgassing model, $(R_{12^\circ C}^{4^\circ C})_{exp}$ and $(R_{12^\circ C}^{20^\circ C})_{exp}$ were compared with $(R_{12^\circ C}^T)_{theo}$, defined in eq 11, in the range of temperature between 2 and 22 °C (dashed line).

Moreover, since the density of champagne does not drastically vary in the range of champagne temperatures investigated in our experiments, since the ambient pressure remains close to 1 bar (i.e., 10^5 N m^{-2}), since the number N of bubble nucleation sites remains the same from one experience to another, and since the distance h traveled by bubbles is the same from one experience to another, CO_2 volume fluxes released by bubbles are finally expected to follow the following scaling law in the case of a given champagne sample poured in the engraved flute:

$$\frac{dV}{dt} \propto \frac{T^{11/3}}{\eta} \tag{9}$$

By replacing in eq 9 the dynamic champagne viscosity by its Arrhenius-like equation given in eq 1, the following scaling law was derived for the temperature dependence of CO_2 volume fluxes released by champagne bubbles collapsing at the champagne surface in tasting conditions:

$$\frac{dV}{dt} \propto \frac{T^{11/3}}{\exp(2806/T)} \tag{10}$$

The previous scaling law teaches us that, for a given champagne sample, CO_2 volume fluxes released by bubbles are strongly champagne temperature-dependent. By arbitrarily choosing a reference temperature of 12 °C (i.e., 285 K), let us build the following theoretical ratio $(R_{12^\circ C}^T)_{theo}$, which compares the CO_2 volume flux outgassing from a given champagne sample at a given temperature T with the CO_2 volume flux outgassing from the same champagne sample at 12 °C (every other parameter being equal under the same operating conditions):

$$(R_{12\,^\circ C}^T)_{theo} = \frac{(dV/dt)_T}{(dV/dt)_{12\,^\circ C}} = \frac{T^{11/3}}{(285)^{11/3}} \times \exp\left(\frac{2806}{285} - \frac{2806}{T}\right) \tag{11}$$

The above-defined theoretical ratio $(R_{12^\circ C}^T)_{theo}$ was plotted in **Figure 7**, as a function of champagne temperature T (between

2 and 22 °C). In order to test the temperature dependence of this first outgassing model based on results obtained over recent years, $(R_{12°C}^{T})_{\text{theo}}$ was also compared with the two experimentally determined ratios, $(R_{12°C}^{4°C})_{\text{exp}}$ and $(R_{12°C}^{20°C})_{\text{exp}}$ (obtained just after pouring, i.e., at $t = 0$), which both compare the average CO$_2$ volume fluxes outgassing from champagne, at 4 and 20 °C, respectively, with the average CO$_2$ volume flux outgassing from champagne at the reference temperature of 12 °C. It is clear from **Figure 7** that a quite good correspondence exists between the theoretical ratio defined in eq 11 and the two experimentally determined ratios, $(R_{12°C}^{4°C})_{\text{exp}}$ and $(R_{12°C}^{20°C})_{\text{exp}}$, respectively. This last figure seems to indicate that the bubbling model given in eq 8, built upon results obtained over recent years, reproduces quite well the influence of champagne temperature on the kinetics of CO$_2$ fluxes outgassing from a flute. Actually, it is worth noting that there are indeed two ways for progressive CO$_2$ losses after the champagne is poured into a flute. CO$_2$ molecules can escape (i) in the form of bubbles and (ii) by diffusion through the free air/champagne interface (i.e., the area of the flute) (1). In the present paragraph, CO$_2$ losses were modeled only through the bubbling way. A complete model taking into account the two ways of CO$_2$ molecule escape is under construction, but goes beyond the scope of the present article.

For the very first time, measurements of CO$_2$ fluxes outgassing from a flute poured with champagne were presented, in tasting conditions, all along the first 10 min following the pouring process. Experiments were performed at three sets of temperature, namely, 4 °C, 12 °C, and 20 °C, respectively. It was clearly demonstrated that the lower the champagne temperature, the lower CO$_2$ volume fluxes outgassing from the flute. Therefore, the lower the champagne temperature, the lower its progressive loss of dissolved-CO$_2$ concentration with time, which constitutes the first analytical proof that low champagne temperatures prolong the drink's chill and helps retains its effervescence. Moreover, in champagne and sparkling wine tasting, the driving force of CO$_2$ desorption from the supersaturated liquid matrix being proportional to its concentration of dissolved CO$_2$ denoted c_L, we proposed a correlation between CO$_2$ volume fluxes outgassing from the flute and c_L. Finally, the contribution of effervescence to the global kinetics of CO$_2$ release was discussed and modeled by the use of several results developed over recent years. On the basis of this bubbling model, the theoretical influence of champagne temperature on CO$_2$ volume fluxes outgassing from a flute was discussed and found to be in good accordance with our experimental results. In the near future, we plan to test the influence of glass design on the kinetics of CO$_2$ release from champagne or sparkling wine by, for example, the use of glasses showing various glass shapes and engravement conditions. We will also investigate the influence of other various parameters of the liquid medium on the kinetics of CO$_2$ release, such as its viscosity, and liquid level in the glass.

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