ARTICLE

Losses of Dissolved CO₂ Through the Cork Stopper during Champagne Aging: Toward a Multiparameter Modeling

Gérard Liger-Belair*,†,‡ and Sandra Villaume†

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

‡Equipe Effervescence, Groupe de Spectrométrie Moléculaire et Atmosphérique (GSMA), UMR CNRS 6089, UFR Sciences Exactes et Naturelles, B.P. 1039, 51687 Reims Cedex 2, France

ABSTRACT: Measurements of dissolved CO_2 concentrations from Champagne bottles initially holding the same CO_2 level after having been elaborated (close to 11.5 g L^{-1}), but having experienced different periods of aging after having been corked with natural cork stoppers, were done. Losses of dissolved CO_2 close to 3.5 g L^{-1} experienced by the oldest Champagne samples aged for about 75 months were reported. This very significant loss of dissolved CO_2 was logically interpreted as a consequence of the continuous diffusion of gaseous CO_2 through the pores of the cork stopper. By combining the diffusion principle through a porous medium with Henry's law (which links the solubility of a gas species in a liquid medium with its partial pressure in the vapor phase), a multiparameter model was built that provides the dissolved CO_2 content found in Champagne during its whole aging period. Both Champagne temperature and bottle volume were found to be key parameters with regard to the kinetics of CO_2 losses through the cork.

KEYWORDS: Champagne, dissolved CO_2, cork stopper, diffusion, Champagne aging

■ INTRODUCTION

From a strictly chemical point of view, Champagne and sparkling wines elaborated according to the *méthode traditionnelle* are multicomponent hydroalcoholic systems supersaturated with CO_2 dissolved gas molecules formed together with ethanol during the second fermentation process, called *prise de mousse* (promoted by adding a certain amount of sugar inside bottles filled with a base wine and sealed with a cap). 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.[1,2] 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 therefore hold a concentration of dissolved CO_2 proportional to the level of sugar added to promote this second fermentation (see, for example, ref 3 and references cited therein). The concentration of dissolved CO_2 in Champagne (in grams per liter) is roughly equivalent to half of the concentration of sugar (in grams per liter) added into the base wine to promote the *prise de mousse*. Traditionally, 24 g/L sugar is added in the base wine to promote the *prise de mousse*. Therefore, a standard Champagne holds close to 12 g/L dissolved CO_2 molecules after the *prise de mousse*, that is, about 9 g per each standard 75 cL bottle.[1-3]

After the *prise de mousse*, Champagne ages in a cool cellar for at least 15 months to develop its so-called bouquet. Bottles then undergo disgorging. Caps are removed to remove the sediment of dead yeast cells. Specific liquor, which consists of a mixture of sugar and old wines, is then added to replace the bit of wine lost during disgorging. A bit of dissolved CO_2 is also inevitably lost at this step, but bottles are quickly corked with traditional or technical cork stoppers to prevent an excessive loss of CO_2. Experiments with early disgorged Champagne samples were done recently, and the characteristic concentration of dissolved CO_2 inside the bottle was found to be of the order of 11.5 g/ L.[4-6]

In Champagne and sparkling wine tasting, the concentration of dissolved CO_2 is a parameter of wide importance because it directly affects the four following sensory properties: (i) the frequency of bubble formation in the glass;[7-9] (ii) the growth rate of rising bubbles;[7-9] (iii) the mouthfeel, that is, the mechanical action of collapsing bubbles as well as the chemosensory excitation of nociceptors in the oral cavity (via the conversion of dissolved CO_2 to carbonic acid);[10-12] and (iv) the nose of Champagne, that is, its so-called bouquet.[13,14] Consequently, keeping the dissolved CO_2 molecules as long as possible inside the bottle during aging is a challenge of wide importance for Champagne wine elaborators.

Natural cork stoppers are commonly used for Champagne corking because of their remarkable impermeability to liquid and air (thus delaying wine from oxidation), elasticity, resiliency combined with compressibility, and chemical inertness.[15,16] Nevertheless, despite all of these remarkable properties, the natural cork is not completely hermetic with regard to gas transfers. Cork is a porous material. Gas species are therefore able to slowly diffuse through the cork along their respective inverse partial pressure gradients. In the case of a corked Champagne bottle, the huge difference in CO_2 partial pressure between the headspace (close to 6 bar at 12 °C) and the ambient air (of the order of only 0.0004 bar)

Table 1. Various Periods of Aging, Corresponding Dissolved CO_2 Concentrations, Lengths of Cork, and Cross Sections of the Bottle Neck Found in Both Champagne Types[a]

type of Champagne	period of aging before uncorking, t_i (months)	concentration of dissolved CO_2 in the corresponding wine, c_i^W (g L^{-1})	length of cork penetrating into the bottleneck δ (cm)	cross section of the bottleneck (equivalent to the cork section), S (cm^2)
YC	2	11.6 ± 0.3	2.6 ± 0.1	2.4
OC	75 ± 1	8.2 ± 0.3	2.5 ± 0.1	2.4

[a] Uncertainty of ± 1 month concerning the period of aging of old samples is due to the fact that the disgorging and bottling of those samples was spread over 2 months. To enable a statistical treatment, six bottles of each Champagne type were used to determine an average dissolved CO_2 concentration corresponding to each Champagne type (with standard deviations corresponding to the root-mean-square deviations provided by the six samples).

forces CO_2 molecules to slowly diffuse through the cork. Therefore, during its aging period, a corked Champagne bottle progressively loses its gaseous CO_2 content. Moreover, Champagne inevitably loses its dissolved CO_2 content as well (because gaseous CO_2 and dissolved CO_2 both experience equilibrium through Henry's law).

In this paper, losses of dissolved CO_2 from Champagne bottles having experienced significant periods of aging were reported. This very significant loss of dissolved CO_2 experienced by samples aged for several years was logically interpreted as a consequence of a continuous diffusion of gaseous CO_2 through the cork's pores. By combining the diffusion principle through a porous media with Henry's law (which links the solubility of a gas species in a liquid medium with its partial pressure in the vapor phase), a multiparameter model was built that describes the dissolved CO_2 content found in Champagne during its whole aging period.

MATERIALS AND METHODS

Champagne Samples. Standard commercial Champagne wines, provided by our industrial partner (Champagne Vranken-Pommery, Reims, France), were used for this set of experiments. Champagne wines elaborated in standard bottles (75 cL) were chosen. Bottles having experienced two significantly different periods of aging (after having been corked with natural cork stoppers) were chosen and compared to each other with regard to their dissolved CO_2 contents, namely, (1) a young Champagne wine, recently elaborated (vintage 2007, disgorged and corked in 2009), stored in a cool cellar at 12 °C since, and referred to as YC in the following; and (2) an older Champagne wine, elaborated in the late 1990s (vintage 1998, disgorged and corked in 2001), stored in a cool cellar at 12 °C since, and referred to as OC in the following.

It is worth noting that both Champagne types were elaborated with 24 g/L sugar added in the base wine to promote the *prise de mousse*, so that the same amount of about 12 g/L of CO_2 was initially dissolved into both Champagne types. This was definitely a crucial condition to correctly evaluate the loss of dissolved CO_2 during the aging process.

Measuring Concentrations of CO_2 Dissolved in Champagne Samples. Concentrations of dissolved CO_2 in the various Champagne samples were determined using carbonic anhydrase (labeled C2522 Carbonic Anhydrase Isozyme II from bovine erythrocytes and provided from Sigma-Aldrich). This is the official method recommended by the OIV (namely, the International Office of Vine and Wine) for measuring the dissolved CO_2 concentration in Champagne and sparkling wines.[17] This method is thoroughly detailed in a recent paper by Liger-Belair et al.[4] The concentration of dissolved CO_2 in bottles was carried out immediately after uncorking the bottle (to prevent a loss of dissolved CO_2 due to the progressive and ineluctable diffusion once a bottle is uncorked). To enable a statistical treatment, six bottles of each Champagne type were used to determine an average dissolved CO_2 concentration corresponding to each Champagne type (with standard deviations corresponding to the root-mean-square deviations provided by the six samples).
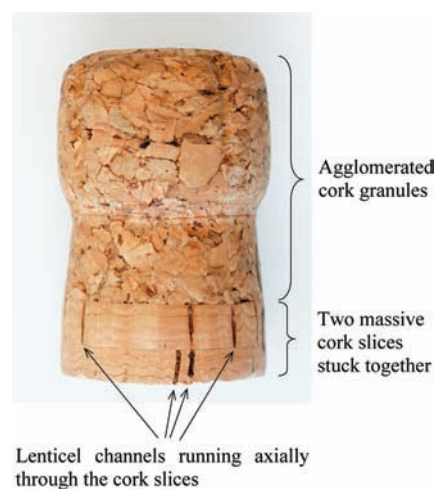

Figure 1. Side view of a natural cork stopper freshly uncorked from a standard "young" Champagne bottle; it clearly appears that it is composed of two different parts: (i) a upper part composed of agglomerated cork granules and (ii) a lower part made of two massive cork slices stuck together (where several lenticel channels clearly appear).

RESULTS AND DISCUSSION

Concentrations of Dissolved CO_2 Found Inside Both Champagne Types. Table 1 compiles the concentrations of dissolved CO_2 found in both Champagne types. It is worth noting that the average concentration of dissolved CO_2 in the young Champagne is quite close to the one found recently in other Champagne wines recently elaborated, by use of the same technique,[4−6] and through a nondestructive and unintrusive method based on a nuclear magnetic resonance technique.[18] As clearly displayed in Table 1, the average concentration of dissolved CO_2 found in the old Champagne is significantly lower than the one found in the young one (despite the same initial amount of dissolved CO_2 after the *prise de mousse*). This observation betrays the fact that CO_2 dissolved molecules progressively escape from Champagne during aging.

Actually, the thick glass of a Champagne bottle being perfectly hermetic with regard to gas exchanges, the cork stopper is therefore logically the only way for gaseous CO_2 molecules to escape from inside the bottle to reach the ambient air. The aim of the following paragraph is to propose a theoretical model that links the concentration of dissolved CO_2 in a Champagne bottle during aging with several physicochemical and geometrical parameters of the wine, cork, and bottle itself.

Toward a Multiparameter Modeling of the Dissolved CO_2 Concentration during Aging. During aging, gaseous CO_2 molecules can escape from inside the bottle to reach the ambient air by pushing their way through two distinct "pathways", namely,
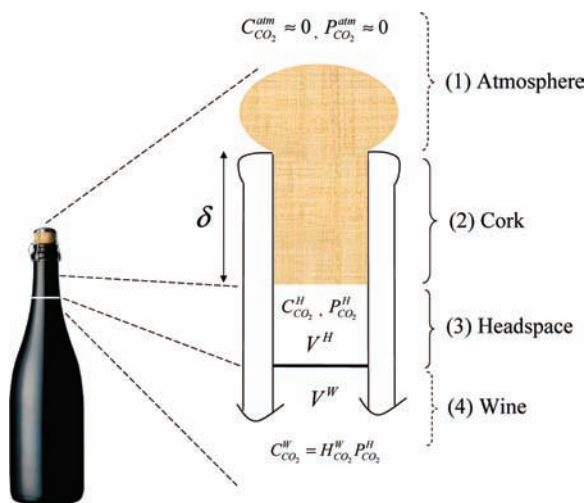
Figure 2. Scheme of the bottleneck and compilation of the various cork, bottle, and wine parameters (see Abbreviations Used for more details about each parameter).

(i) through the porous cork bulk or (ii) through the cork/bottleneck interface, that is, the frontier between the cork and the glass. Moreover, the traditional cork stopper for Champagne and sparkling wines is neither a homogeneous nor an isotropic "object". It is composed of two distinct parts. The mushroom-shaped upper part of the cork is composed of agglomerated cork granules, whereas its lower part is made of two massive cork slices stuck together, as illustrated in the photograph displayed in Figure 1. These two distinct cork parts certainly present various mechanical and porous properties. For example, because the lower part of the cork is made of two massive cork slices, several lenticels (i.e., quasi cylindrical channels composed of dead cell walls and intercellular spaces) cross the slices axially and certainly provide a specific way for gas diffusion (see Figure 1). Nevertheless, the diameter and number of lenticels significantly varies from one type of cork to another. In the paper by Lequin et al., scanning electron microscope pictures of the cork cell structure are displayed, including several views of lenticels.[19] For all of the aforementioned reasons, it seems very difficult to propose a model that depicts the details of each mechanism by which CO_2 molecules progressively diffuse through the cork during aging. This is why we have decided to adopt a more global approach, which simply considers the natural cork stopper as a nonhermetic closure that enables the gas diffusion process (no matter how and where CO_2 molecules push their way at the molecular scale, in a first step).

Molecular diffusion is actually the mechanism behind the diffusion of gas species through a porous medium. Molecular diffusion classically obeys the first Fick's law, which stipulates that the flux of a given gas species through a porous material is proportional to its concentration gradient along the porous material, as

$$J = -D \times \nabla c \tag{1}$$

where D is the diffusion coefficient of the gas species in the porous material (including the various pathways by which CO_2 molecules can diffuse through the cork in the present work, namely, the cork bulk and the cork/bottleneck interface) and ∇c is its gradient concentration along the porous material (i.e., from

the headspace under the cork to the open atmosphere in the present case).

By assuming a linear gradient of gaseous CO_2 between the headspace under the cork and the open atmosphere, ∇c may be rewritten as $\Delta c/\delta$, with δ being the length of the cork and Δc being the concentration difference in gaseous CO_2 between the headspace under the cork and the open atmosphere. The number n of CO_2 moles that cross the porous cork per unit of time is therefore ruled by

$$\frac{dn}{dt} = \iint\limits_{\text{cork section}} J \times dS \approx -DS\frac{\Delta c}{\delta} \tag{2}$$

where S is the cross section of the cork that separates the headspace of the Champagne bottle from the open atmosphere (equivalent indeed to the cross section of the bottleneck).

The scheme displayed in Figure 2 compiles the various physicochemical and geometrical pertinent parameters of the model. Because the present work specifically deals with the dissolved CO_2 concentration found in Champagne samples, and to facilitate the reading of the paper, the subscript CO_2 found in the formulas will be omitted in the following.

The total number of CO_2 moles, denoted n^{total}, found in the whole bottle is the sum of the number of dissolved CO_2 moles found in the Champagne bulk, denoted n^W, and the number of gaseous CO_2 moles found in the headspace under the cork, denoted n^H. Moreover, as shown in Figure 2, the dissolved CO_2 concentration found in the Champagne bulk, denoted c^W, and the pressure of gaseous CO_2 under the cork, denoted P^H, experience equilibrium through the so-called Henry's law. Finally, by considering the gaseous CO_2 in the headspace as an ideal gas, the following system of equations may be written

$$\begin{cases} n^{\text{total}} = n^W + n^H \\ c^W = H^W P^H \\ P^H V^H = n^H RT \end{cases} \tag{3}$$

with R being the ideal gas constant ($8.31\ \text{J K}^{-1}\ \text{mol}^{-1}$), T being the absolute temperature (in K), V^H being the volume of the gaseous headspace under the cork, and V^W being the volume of the Champagne liquid phase.

By replacing in the preceding system c^W by n^W/V^W and P^H by $n^H RT/V^H$, the eq 3 may be rewritten as follows:

$$\begin{cases} n^{\text{total}} = n^W + n^H \\ n^W = V^W \dfrac{H^W RT}{V^H} n^H \end{cases} \tag{4}$$

The total number of CO_2 moles that crosses the cork is ruled by eq 2, with $\Delta c \approx c^H \approx n^H/V^H$ (because the concentration of gaseous CO_2 in the open atmosphere is negligible in comparison with that found in the headspace under the cork). Therefore

$$\frac{dn^{\text{total}}}{dt} = \frac{dn^W}{dt} + \frac{dn^H}{dt} \approx -DS\frac{n^H}{\delta V^H} \tag{5}$$

By combining systems of eqs 4 and 5 and by replacing n^H by $P^H V^H/RT$, eq 5 transforms as

$$\left(1 + \frac{V^W H^W RT}{V^H}\right)\frac{dP^H}{dt} + \frac{DS}{\delta V^H}P^H = 0 \tag{6}$$

The partial pressure of gaseous CO_2 in the headspace under the cork is finally ruled by the differential equation

$$\frac{dP^H}{dt} + \frac{DS}{\delta(V^H + V^W H^W RT)} P^H = 0 \qquad (7)$$

which admits the following analytical solution:

$$\begin{cases} P^H(t) \approx P_0^H \exp\left(-\frac{t}{\tau}\right) \\ \tau = \frac{\delta(V^H + V^W H^W RT)}{DS} \end{cases} \qquad (8)$$

τ is the characteristic time scale of an exponential decay law, and P_0^H is the initial partial pressure of gaseous CO_2 in the headspace under the cork.

In the present work, it is worth noting that evaporation of the liquid phase (which may sometimes occur during long aging periods of time) was considered as being negligible, thus keeping the respective volumes of Champagne liquid phase and gaseous headspace constant.

By considering that the concentration of dissolved CO_2 and the partial pressure of gaseous CO_2 in the headspace experience equilibrium all along the aging of Champagne, the concentration of dissolved CO_2 found in the Champagne bulk also follows an exponential decay law with the same characteristic time scale τ:

$$c^W(t) \approx H^W P^H(t) \approx c_0^W \exp\left(-\frac{t}{\tau}\right) \qquad (9)$$

with c_0^W being the initial concentration of dissolved CO_2 found in the Champagne bulk after bottle corking.

It is worth noting that, from eq 9, it becomes indeed possible to access an experimental determination of the diffusion coefficient of the natural cork, in situ. Actually, for both Champagne types, the concentrations of dissolved CO_2 found in the Champagne bulk after uncorking were determined at two steps of the Champagne aging process, respectively, after 2 months and after 75 months of aging (see Table 1). The two periods of aging were denoted t_1 and t_2, respectively. Following eq 9, the concentrations of dissolved CO_2 theoretically found inside the Champagne, after these two distinct periods of aging, may be assessed by

$$\begin{cases} c_1^W \approx c_0^W \exp\left(-\frac{t_1}{\tau}\right) \\ c_2^W \approx c_0^W \exp\left(-\frac{t_2}{\tau}\right) \end{cases} \qquad (10)$$

By dividing c_1^W by c_2^W and by developing the characteristic time scale τ, the exponential decay law may by assessed by

$$\tau \approx \frac{t_2 - t_1}{\ln(c_1^W / c_2^W)} \qquad (11)$$

Finally, by retrieving the theoretical multiparameter determination of τ given in eq 8 and by replacing τ by its expression given in eq 11, the diffusion coefficient D of CO_2 molecules in the natural cork may be indirectly assessed as follows:

$$D = \frac{\delta(V^H + V^W H^W RT) \ln(c_1^W / c_2^W)}{S(t_2 - t_1)} \qquad (12)$$

By replacing each parameter in eq 12 by its numerical expression (see Abbreviations Used for more details), it becomes possible to

determine a reasonable experimental value for the diffusion coefficient of CO_2 molecules in the natural cork (each parameter in eq 12 should be given in the MKSA system)

$$D_{exp}$$
$$= \frac{2.6 \times 10^{-2}(2.5 \times 10^{-5} + 7.5 \times 10^{-4} \times 4.5 \times 10^{-4} \times 8.3 \times 285) \ln(11.6/8.2)}{2.4 \times 10^{-4}(1.9 \times 10^8)}$$
$$\approx 1.6 \times 10^{-10} \text{ m}^2 \text{ s}^{-1} \qquad (13)$$

From the theoretical model built hereafter, and from experimental data about the loss of dissolved CO_2 molecules during aging, it has been possible to indirectly propose an experimental approximation of the diffusion coefficient of CO_2 molecules through the porous cork. This diffusion coefficient is logically much smaller than the one of CO_2 molecules in ambient air, which is $\approx 1.8 \times 10^{-5}$ m^2 s^{-1}, at 0 °C.[20] To the best of our knowledge, this is the first attempt to determine in situ the CO_2 diffusion coefficient through the natural cork (including the various pathways by which CO_2 molecules can diffuse through the cork in the present work, namely, the cork bulk and the cork/bottleneck interface).

Actually, the theoretical model built above enables the examination of the impact of some pertinent parameters on the losses of dissolved CO_2 during aging. The goal of the two following paragraphs is to discuss the theoretical role of both Champagne temperature and bottle volume on the progressive loss of dissolved CO_2 during aging.

Role of Temperature. The temperature appears in the characteristic time scale τ of the exponential decay law given in eq 8. Moreover, it is worth noting that the temperature is also "hidden" in another parameter, which is indeed strongly temperature dependent, namely, the Henry's law constant, denoted H^W, which reflects the solubility of CO_2 in the wine. The temperature dependence of the CO_2 solubility in a typical Champagne wine was already determined, for a wide range of Champagne temperatures.[21] It is well described with a van't Hoff like equation as

$$H^W(T) = H_{298\,K}^W \exp\left[-\frac{\Delta H_{diss}}{R}\left(\frac{1}{T} - \frac{1}{298}\right)\right] \qquad (14)$$

where $H_{298\,K}^W$ is the solubility of CO_2 in a typical Champagne wine at 298 K and ΔH_{diss} is the dissolution enthalpy of CO_2 molecules in the liquid medium (in J/mol). The best fit to experimental data was found with $\Delta H_{diss} \approx 24800$ J/mol and with $H_{298\,K}^W \approx 1.21$ g L^{-1} bar^{-1}, that is, $H_{298\,K}^W \approx 2.75 \times 10^{-4}$ mol m^{-3} Pa^{-1}.

For modeling purposes, the diffusion coefficient of CO_2 molecules in the natural cork is supposed to be slowly temperature dependent (in a "reasonable" range of Champagne aging temperature), and the experimentally approached value determined in the previous paragraph will be used hereafter. The graph displayed in Figure 3 shows the theoretical loss of dissolved CO_2 during Champagne aging, for a standard Champagne bottle stored at three different temperatures, namely, 4, 12, and 20 °C. Following Figure 3, it is clear that the lower the temperature during Champagne aging, the lower the losses of dissolved CO_2 through the cork stopper.

Role of Bottle Volume. Champagne wines can be elaborated, disgorged, and corked in bottles of various shapes and volume content. Most commonly, Champagne is elaborated in three various types of bottle, namely, the standard bottle (with a volume of 75 cL), the magnum bottle (with a volume of 150 cL), and the
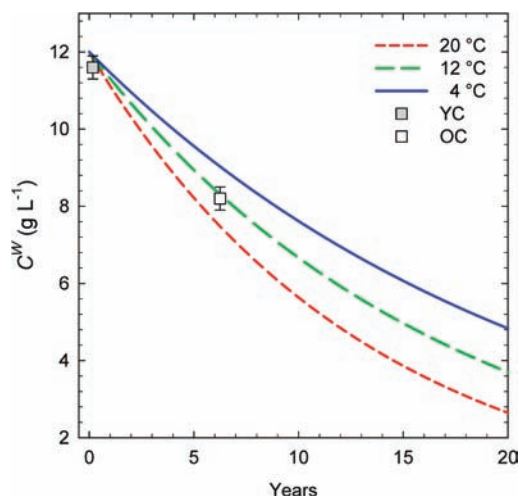
Figure 3. Following eq 9, theoretical concentrations of dissolved CO_2 found in the wine during the aging process (in a standard 75 cL bottle) and for three distinct cellar temperatures, namely, 4, 12, and 20 °C, respectively. Our experimental measurements of dissolved CO_2 found in both Champagne types also appear.

Figure 4. Following eq 9, theoretical concentrations of dissolved CO_2 found in the wine during the aging process (at a given cellar temperature of 12 °C) and for three distinct bottle types, namely, a magnum bottle, a standard bottle, and a half bottle, respectively. Our experimental measurements of dissolved CO_2 found in both Champagne types also appear.

half-bottle (with volume of 37.5 cL). It is worth noting that these three various bottles present indeed different volume contents, but the same headspace volume V^H, as well as similar cork stoppers (in terms of length, δ, and cross section, S). The graph displayed in Figure 4 shows the theoretical loss of dissolved CO_2 during Champagne aging, for the three above-mentioned contents (at a given aging temperature of 12 °C). Following Figure 4, it is clear that the larger the volume content, the lower the losses of dissolved CO_2 through the cork stopper. It is indeed well-known from Champagne connoisseurs that Champagne and sparkling wines age longer and better in magnums than in smaller bottle types.[2,3]

In conclusion, losses of dissolved CO_2 from Champagne bottles, initially holding the same CO_2 level after having been elaborated (close to 11.5 g L^{-1}) but having experienced significant periods of aging, were reported. This very significant loss of dissolved CO_2 (close to 3.5 g L^{-1}) experienced by samples aged for about 75 months after having been corked was logically interpreted as a consequence of the continuous diffusion of gaseous CO_2 through the pores of the cork stopper or through the cork/glass interface. By combining the diffusion principle through a porous medium with Henry's law (which links the solubility of a gas species in a liquid medium with its partial pressure in the vapor phase), a multi-parameter model was built that provides the theoretical dissolved CO_2 content found in a Champagne wine during its whole aging period. Both Champagne temperature and bottle volume were found to be key parameters with regard to the kinetics of CO_2 losses through the cork.

This paper is considered as a first attempt to predict how a bottle of Champagne or sparkling wine progressively loses its dissolved CO_2 content during aging. Nevertheless, it is worth noting that this model considers the diffusion coefficient of gaseous CO_2 through the cork as a constant during aging, which may not be necessarily the case. Actually, despite fantastic resiliency properties, the cork alters as time proceeds. It progressively loses its elasticity, which may in turn modify its porosity and therefore permeability with regard to gas transfers (especially through the cork/glass interface). Moreover, during the aging process, it is also well-known that the pore space of the cork stoppers is progressively invaded by the
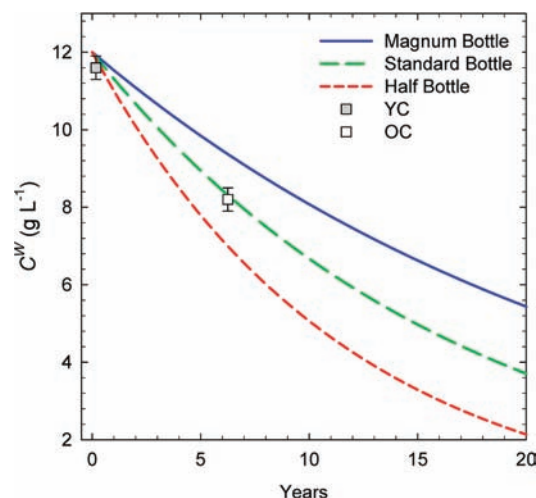
liquid. This phenomenon could also progressively modify the way CO_2 molecules diffuse from inside the bottle to the ambient air, thus modifying in turn the model proposed in this paper. Deeper experimental investigations about the way a cork stopper alters with time in a bottleneck are needed. Such experiments are soon to be conducted along that line.

■ AUTHOR INFORMATION

Corresponding Author
*Phone/fax: + 333 26 91 86 14. E-mail: gerard.liger-belair@univ-reims.fr.

■ ABBREVIATIONS USED

c^W,	dissolved CO_2 concentration found in the Champagne bulk, in mol m^{-3}
c_0^W,	dissolved CO_2 concentration initially found in the Champagne bulk after bottle corking (i.e., at $t = 0$), in mol m^{-3}
D,	diffusion coefficient of gaseous CO_2 molecules through the porous cork, in m^2 s^{-1}
H^W,	Henry's law constant (i.e., solubility of CO_2 molecules in Champagne), in mol m^{-3} Pa^{-1}
n^{total},	total CO_2 found in a Champagne bottle, in mol
n^H,	gaseous CO_2 found in the headspace under the cork, in mol

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n^W,	dissolved CO_2 found in the Champagne bulk, in mol
OC,	old Champagne samples
P^H,	pressure of gaseous CO_2 under the cork, in Pa
P_0^H,	initial pressure of gaseous CO_2 after bottle corking (i.e., at $t = 0$), in Pa
R,	ideal gas constant, 8.31 J K^{-1} mol^{-1}
S,	cross section of the cork stopper (equivalent indeed to the cross section of the bottleneck), in m^2
t,	time, in s
T,	temperature, in K
V^H,	volume of the gaseous headspace under the cork, in m^3
V^W,	volume of the Champagne liquid phase inside the bottle, in m^3
YC,	young Champagne samples
δ,	length of cork penetrating into the bottleneck in m
τ,	characteristic time of the exponential decay law, in s

■ REFERENCES

(1) Liger-Belair, G.; Polidori, G.; Jeandet, P. Recent advances in the science of champagne bubbles. *Chem. Soc. Rev.* **2008**, *37*, 2490–2511.

(2) Liger-Belair, G.; Rochard, J. *Les vins effervescents* ; Dunod: Paris, 2008.

(3) Duteurtre, B. *Le Champagne: De la Tradition à la Science*; Lavoisier: Paris, France, 2010.

(4) Liger-Belair, G.; Villaume, S.; Cilindre, C.; Jeandet, P. Kinetics of CO_2 fluxes outgassing from champagne glasses in tasting conditions: the role of temperature. *J. Agric. Food Chem.* **2009**, *57*, 1997–2003.

(5) Liger-Belair, G.; Villaume, S.; Cilindre, C.; Polidori, G.; Jeandet, P. CO_2 volume fluxes outgassing from champagne glasses in tasting conditions: flute versus coupe. *J. Agric. Food Chem.* **2009**, *57*, 4939–4947.

(6) Liger-Belair, G.; Bourget, M.; Villaume, S.; Jeandet, P.; Pron, H.; Polidori, G. On the losses of dissolved CO_2 during champagne serving. *J. Agric. Food Chem.* **2010**, *58*, 8768–8775.

(7) Liger-Belair, G.; Marchal, R.; Vignes-Adler, M.; Robillard, B.; Maujean, A.; Jeandet, P. Study of effervescence in a glass of champagne: frequencies of bubble formation, growth rates and velocities of rising bubbles. *Am. J. Enol. Vitic.* **1999**, *50*, 317–323.

(8) Liger-Belair, G. Nucléation, ascension et éclatement d'une bulle de champagne. *Ann. Phys. (Paris)* **2006**, *31*, 1–133.

(9) Liger-Belair, G.; Parmentier, M.; Jeandet, P. Modeling the kinetics of bubble nucleation in champagne and carbonated beverages. *J. Phys. Chem. B* **2006**, *110*, 21145–21151.

(10) Dessirier, J.-M.; Simons, C. T.; Carstens, M. L.; O'Mahony, M.; Carstens, E. Psychophysical and neurobiological evidence that the oral sensation elicited by carbonated water is of chemogenic origin. *Chem. Senses* **2000**, *25*, 277–284.

(11) Carstens, E.; Carstens, M. L.; Dessirier, J.-M.; O'Mahony, M.; Simons, C. T.; Sudo, M.; Sudo, S. It hurts so good: oral irritation by spices and carbonated drinks and the underlying neural mechanisms. *Food Qual. Pref.* **2002**, *13*, 431–443.

(12) Chandrashekar, J.; Yarmolinsky, D.; von Buchholtz, L.; Oka, Y.; Sly, W.; Ryba, N.; Zuker, C. The taste of carbonation. *Science* **2009**, *326*, 443–445.

(13) Priser, C.; Etievant, P. X.; Nicklaus, S.; Brun, O. Representative Champagne wine extracts for gas chromatography olfactometry analysis. *J. Agric. Food Chem.* **1997**, *45*, 3511–3514.

(14) Tominaga, T.; Guimbertau, G.; Dubourdieu, D. Role of certain volatile thiols in the bouquet of aged Champagne wines. *J. Agric. Food Chem.* **2003**, *51*, 1016–1020.

(15) Silva, S.; Sabino, M.; Fernandes, V.; Correlo, V.; Boesel, L.; Reis, R. Cork: properties, capabilities and applications. *Int. Mater. Rev.* **2005**, *50*, 345–365.

(16) Pereira, H. *Cork: Biology, Production and Uses*; Elsevier: Amsterdam, The Netherlands, 2007.

(17) Dioxyde de Carbone Ref MA-F-AS314-01-DIOCAR. In *Recueil des Méthodes Internationales d'Analyses des Boissons Spiritueuses, des Alcools et de la Fraction Aromatique des Boissons*; Office International de la Vigne et du Vin: Paris, France, 1994.

(18) Autret, G.; Liger-Belair, G.; Nuzillard, J.-M.; Parmentier, M.; Dubois de Montreynaud, A.; Jeandet, P.; Doan, B. T.; Beloeil, J.-C. Use of magnetic resonance technique for the investigation of the CO_2 dissolved in champagne and sparkling wines: a non-destructive and unintrusive method. *Anal. Chim. Acta* **2005**, *535*, 73–78.

(19) Lequin, S.; Chassagne, D.; Karbowiak, T.; Gougeon, R.; Brachais, L.; Bellat, J. P. Adsoprtion equilibria of water vapor on cork. *J. Agric. Food Chem.* **2010**, *58*, 3438–3445.

(20) Massman, W. A review of the molecular diffusivities of H_2O, CO_2, CH_4, CO, O_3, SO_2, NH_3, N_2O, NO, and NO_2 in air, O_2, and N_2 near STP. *Atmos. Environ.* **1998**, *32*, 1111–1127.

(21) Liger-Belair, G. The physics and chemistry behind the bubbling properties of champagne and sparkling wines: a state-of-the-art review. *J. Agric. Food Chem.* **2005**, *53*, 2788–2802.