JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY

Visualization of Mixing Flow Phenomena in Champagne Glasses under Various Glass-Shape and Engravement Conditions

GÉRARD LIGER-BELAIR,*,† JEAN-BAPTISTE RELIGIEUX,‡ STÉPHANE FOHANNO,‡ MARIE-ANNE VIALATTE,§ PHILIPPE JEANDET,† AND GUILLAUME POLIDORI‡

Laboratoire d'Œnologie et Chimie Appliquée, UPRES EA 2069, URVVC, Faculté des Sciences de Reims, B.P. 1039, 51687 Reims Cedex 2, France, Laboratoire de Thermomécanique, UTAP EA 3802, Faculté des Sciences de Reims, B.P. 1039, 51687 Reims Cedex 2, France, and Centre de Recherche Appliquée et de Transfert d'Expertise (CREATE), rue du Quartier de Cavalerie, 62500 Saint Omer, France

For the very first time, a classical flow visualization technique was used to capture the fluid motion in traditional flutes and coupes poured with champagne. It was found that glasses engraved around their axis of symmetry produce a rising gas column along the vertical glass axis that induces, in turn, steady state recirculating flow regions. In the case of the classical engraved champagne flute, the whole domain of the liquid phase is homogeneously mixed, whereas in the case of the engraved champagne coupe, the recirculating flow region does not occupy the whole volume in the glass. In the engraved coupe, a dead-zone of very low motion was identified, which inhibits the formation of the collar at the glass edge. Our results finally strongly suggest that the glass-shape and engravement conditions should likely have a strong impact on champagne tasting by modifying the kinetics of release of carbon dioxide molecules and aromatic volatile organic compounds from the liquid medium.

KEYWORDS: Champagne; sparkling wines; carbonated beverages; effervescence; bubble nucleation; engravement; laser impact; mixing flow phenomena; convection; flavor release; champagne tasting

INTRODUCTION

In champagne, sparkling wines, and beers, carbon dioxide molecules in excess form together with ethanol when yeast ferment sugars. They are responsible for producing gas bubbles as soon as the bottle is uncorked. In soda drinks and most of fizzy waters, industrial carbonation is the source of effervescence (*1*).

Since the time of the benedictine monk Dom Pierre Perignon (1638−1715), champagne has been the wine of celebration. This fame is undoubtedely largely linked to the elegance of its effervescence and foaming properties (*1*). Critics judge champagne and sparkling wines by, among other qualities, its bubbling behavior. The quality of the product is often related to the size of bubbles formed in the flute. Small bubbles rising slowly through the liquid are usually much preferred to large bubbles. Bubbles formed in the glass are also responsible for the aspect of the foam ring on the liquid surface, the so-called collerette, which is also an important feature of this product. But, even if there is no evidence yet to believe that bubbles confer any other sensory advantage to the wine, it is often recognized that bubbles play a major role in the assessment of champagne and sparkling wines. This is the reason why considerable efforts have been conducted the past few years to better illustrate, detect, understand, and finally control each and every parameter involved in the bubbling process. Generally speaking, effervescence in a glass of champagne or sparkling wine may have two distinct origins. It can be (i) natural or (ii) artificial.

(i) Natural effervescence is related to bubbles nucleated from a glass that has not experienced any specific surface treatment. Closer inspection of such glasses poured with champagne and sparkling wines recently revealed that most of the bubble nucleation sites were found to be located on preexisting gas cavities trapped inside hollow and roughly cylindrical cellulose fiber-made structures on the order of 100 μm long with a cavity mouth of several micrometers (*2*−*6*). These fibers are released from the surrounding air, or from the towel used during the wiping process. Fibers probably adhere on the flute wall due to electrostatic forces (especially if the glass or the flute is vigorously wiped by a towel). A typical fiber acting as a bubble nucleation site is displayed in **Figure 1**. Flutes that have been cleaned with a towel before serving show an excess of bubble nucleation sites and therefore an excess of effervescence (*7*). Therefore, there is a substantial variation concerning the natural

* Corresponding author. Tel.: 00 (33)3 26 91 86 14; fax: 00 (33)3 26 91 33 40; e-mail: gerard.liger-belair@univ-reims.fr.
† Laboratoire d'Œnologie et Chimie Appliquée, Faculté des Sciences de Reims.
‡ Laboratoire de Thermomécanique, Faculté des Sciences de Reims.
§ Centre de Recherche Appliquée et de Transfert d'Expertise.

10.1021/jf062973+ CCC: $37.00
Published on Web 01/17/2007

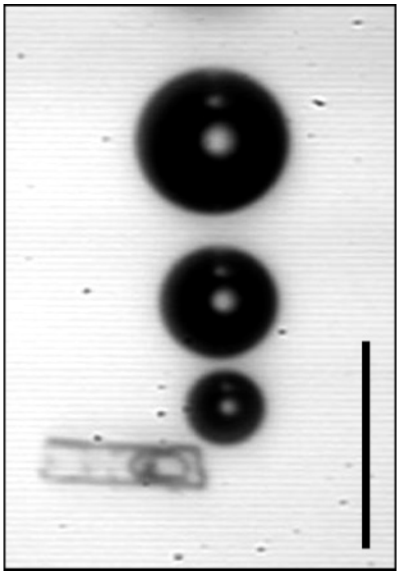

Figure 1. Typical cellulose fiber acting as a bubble nucleation site on the inner wall of a glass poured with champagne, as seen through the microscope objective of a high-speed video camera (see ref *2* for example). This type of bubble nucleation process produces effervescence that is denoted as natural. Bar = 100 μm.

effervescence between flutes depending on how the flute was cleaned and how and where it was left before serving. Very recently, a mathematical model has been derived that reproduces the kinetics of bubbling from cellulose fibers (*8*).

(ii) Artificial effervescence is related to bubbles nucleated from glass imperfections intentionally made by the glassmaker to eventually replace a deficit of natural nucleation sites. Actually, it has been known for decades that bubbles may arise from microscratches on the glass wall (*9*, *10*). Those microscratches trap tiny air pockets when champagne is poured into the glass (as cellulose fibers do). The mechanism of bubble release from cellulose fibers or microscratches has already been described in previous papers (for a review, see i.e., ref *5* and references therein). Those microscratches on a glass or a flute can be done by essentially two techniques: sandblast or laser engraving. A rendering of such microscratches releasing bubbles at the bottom of a champagne flute is displayed in **Figure 2**.

From a 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; beers and sodas would be definitely flat. However, the role of effervescence is not only aesthetic. Actually, bubbles bursting at the liquid surface radiate a cloud of hundreds of tiny droplets every second, as shown in a previous paper (*11*). Those tiny droplets partly evaporate, thus accelerating the transfer of the numerous aromatic volatile organic compounds above the liquid surface in comparison with a flat wine, for example (*1*). Furthermore, effervescence is also believed to play another major role concerning flavor release and gas discharge in glasses poured with champagne. Actually, the continuous flow of ascending bubbles through the liquid strongly modifies the mixing and convection conditions of the liquid medium. In turn, the gas discharge from the liquid surface may be considerably accelerated, as well as the release of the numerous volatile organic compounds that strongly depend on the mixing flow conditions of the liquid medium. Up to now, and to the best of our knowledge, glassmakers chose to engrave some flutes and coupes to increase effervescence and paliate a deficit of natural bubble nucleation sites but without anticipating

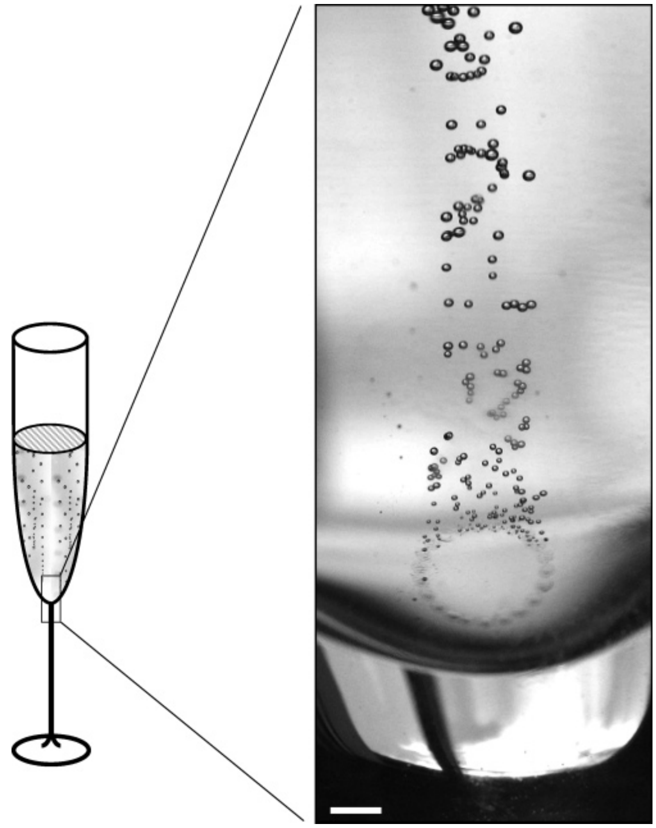

Figure 2. At the bottom of this flute, on its axis of symmetry, the glassmaker has engraved a small ring. Bubbles are seen generated from these artificial microscratches in the form of a vertical bubbles column. This type of bubble nucleation process produces effervescence that is denoted as artificial. Bar = 1 mm. Reprinted with permission from ref *6*.

at all the impact on the kinetics of gas discharge and flavor release from the liquid medium.

The aim of this work based on a classical flow visualization technique is threefold: (i) to visualize and describe for the very first time the mixing flow phenomena in glasses poured with champagne, (ii) to underscore the impact of glass-shape and engravement conditions on the mixing flow phenomena, and (iii) to suggest an impact of glass-shape and engravement on the behavior of bubbles at the free surface, as well as a likely impact on flavor and gas release from the liquid medium.

MATERIALS AND METHODS

A classical Champagne wine holding about 10 g/L of CO_2-dissolved molecules was used for this set of experiments. Some physicochemical parameters of the champagne were already determined at 20 °C, with a sample of champagne first degassed (*3*). The static surface tension of champagne γ was found to be on the order of 47 mN/m, its density ρ was measured and found to be 998 kg/m^3, and its dynamic viscosity η was found to be on the order of 1.5×10^{-3} kg/m/s.

Experiments were performed at room temperature (20 $\pm$ 2 °C). Experiments have been conducted for several commercial champagne glass models, each model being engraved or not. What champagne flow visualizations have indicated is that similar major conclusions can be drawn whatever the glass model. So, for the sake of brevity, only two different and opposite-shape glass models are presented, namely, a slender and elongated one, with a deep tapered bowl (the traditional champagne flute), and another one, with a shallow bowl of widened form (the traditional champagne goblet or coupe). To engrave flutes and coupes and to force artificial effervescence from the glass bottom surface, the glassmaker used a laser beam impact technique according

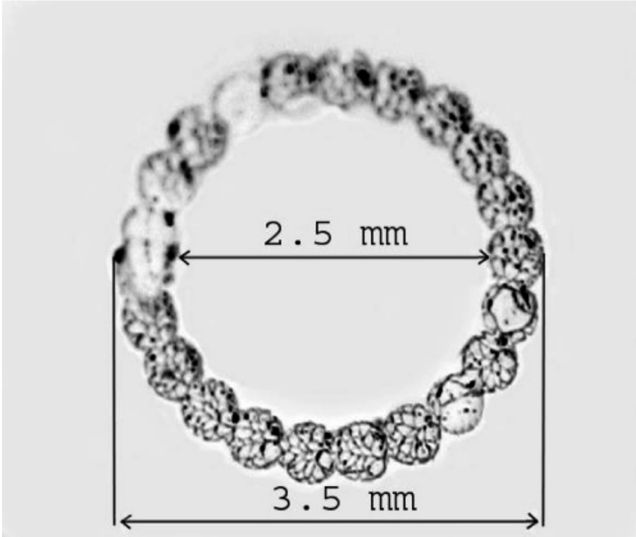

Figure 3. Photographic detail of the small ring engraved at the bottom of the commercial flutes and coupes provided by ARC International. This small ring is a 3.5 mm circle-shaped structure obtained by laser engravement and where each point of impact has a diameter of about 400 μm.

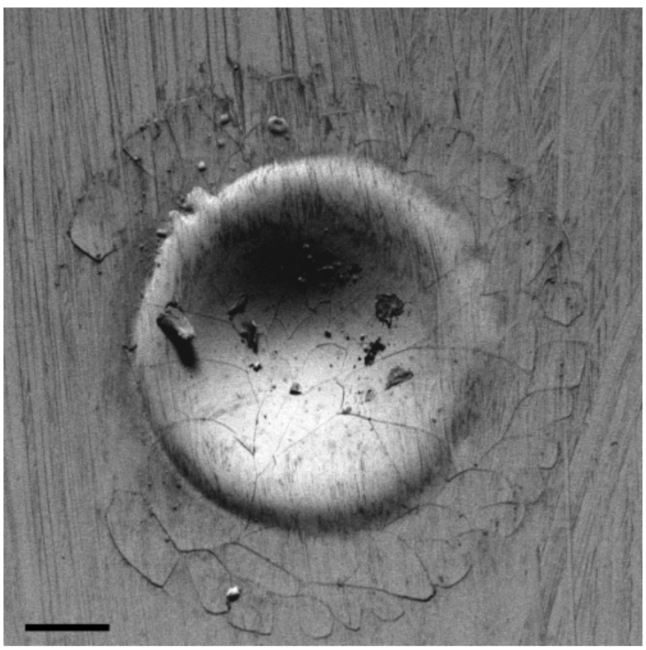

Figure 4. Point of impact viewed through a scanning electron microscope. Bar = 100 μm.

to a circular multipoint distribution whose detail is revealed in **Figure 3**. The point of impact has also been observed through a scanning electron microscope (cf. **Figure 4**).

Classical tracer techniques as dye emission are not suitable for such experiments in close confined domains due to their poor stability behavior against mixing with the surrounding fluid (*12*). It is the reason why the particle-streak technique (*13*) has been chosen for the experiments involving the seed of solid Rilsan particles in the working champagne fluid with a volume fraction condition of $\sim 1.3 \times 10^{-4}$. These spherical particles are neutrally buoyant (75 μm < diameter < 150 μm; $\rho = 1.06$ g/cm^3) and exhibit a high degree of reflectivity when illuminated by a 2 mm thick laser sheet (cf. **Figure 5**). Moreover, Rilsan particles were found to be completely neutral with regard to bubble formation (this was definitely a crucial condition for the feasibility of the present work). The planar laser sheet was built from an argon laser source whose incident beam crosses spherical and cylindrical optical lenses. The particles were initially and carefully introduced into the bottle before the champagne was poured in the glass. After pouring the champagne into the glass, the flow was observed at a normal angle to the light screen, and lighted particles were recorded by a camera with a suitable ($t = 2$ s) exposure time to obtain instantaneous streak fields leading to information on both flow features and kinematics. To palliate the ineluctable chaotic flow behavior inside the glass following the pouring process and to ensure the reproducibility of experiments, a 1 min wait was required and also found to be sufficient enough to reach a quasi-steady state of motion for engraved glasses.

RESULTS AND DISCUSSION

Visualization of the Mixing Flow Patterns. To examine how the mixing flow process occurs within the various glasses, we present in **Figure 6** flow visualizations for the traditional champagne flute, under engravement conditions or not. To enable a better comparison between flow visualization into the various glasses, it is pointed out that pictures have been taken at the same $t = 60$ s time after champagne wine was poured into each glass. To have a better understanding of fluid motion mechanisms, corresponding streamline patterns have been reported in **Figure 7**.

Strong differences appear in the flow behavior according to whether the glass has sustained a specific surface treatment or not. In the case of the traditional champagne flute without surface treatment, the flow is asymmetric in the visualization section and presents large-scale eddies, whose number varies from an experiment to another. The streamline patterns drawn in **Figure 7a** reveal a three-dimensional (3-D) behavior of the flow materialized, for example, by the presence of a spiral in shape focus. Sequences of the flow (data not shown) indicate that the highly rotational viscous flow cells evolve in time, changing in size and location according to an arbitrary scheme. A contrario, using an engraved glass exhibits a steady state of fluid motion reached ~30 s after the glass is poured. Because of the high degree of reflectivity of bubbles, one clearly observes the formation of a rising gas column along the vertical glass axis from the treated bottom surface up to the free surface of the beverage. Consequently, a drive process of the surrounding fluid occurs to generate two large vortices in the vertical section whose rotating wises are mentionned in **Figure 7b**. These cells are located outside of the rising bubbles close to the walls of the glass. Because this gas column acts like a continuous swirling-motion generator within the glass, the flow structure exhibits a two-dimensional (2-D) behavior with an axisymmetrical geometry. It is mentioned that in this case the whole domain of the liquid phase is homogeneously mixed. Prof. Richard Zare and colleagues from Stanford University have recently reported that, in Guinness beer, tiny bubbles could eventually sink on the sides of the container while rising in the center during settling (*14*, *15*). Actually, tiny beer bubbles are not sufficiently buoyant to overcome the vigorous recirculation directed downward by the edges of the glass. However, it is worth noting that in flutes poured with champagne and sparkling wines, bubbles are significantly larger than those in beer (*1*) and therefore sufficiently buoyant to rise despite this recirculating flow directed downward by the edges of the flute (i.e., the velocity of ascending bubbles remains higher than the velocity of the fluid recirculating downward along the flute's walls).

To complete the previous observed trends, we present in **Figure 8** the resulting flow in the engraved traditional champagne coupe, much wider but shallower than the traditional champagne flute. It can be seen that, as for the previous glass model, the rising carbon dioxide bubble column causes the main fluid to move inside the glass. Nevertheless, two distinctive

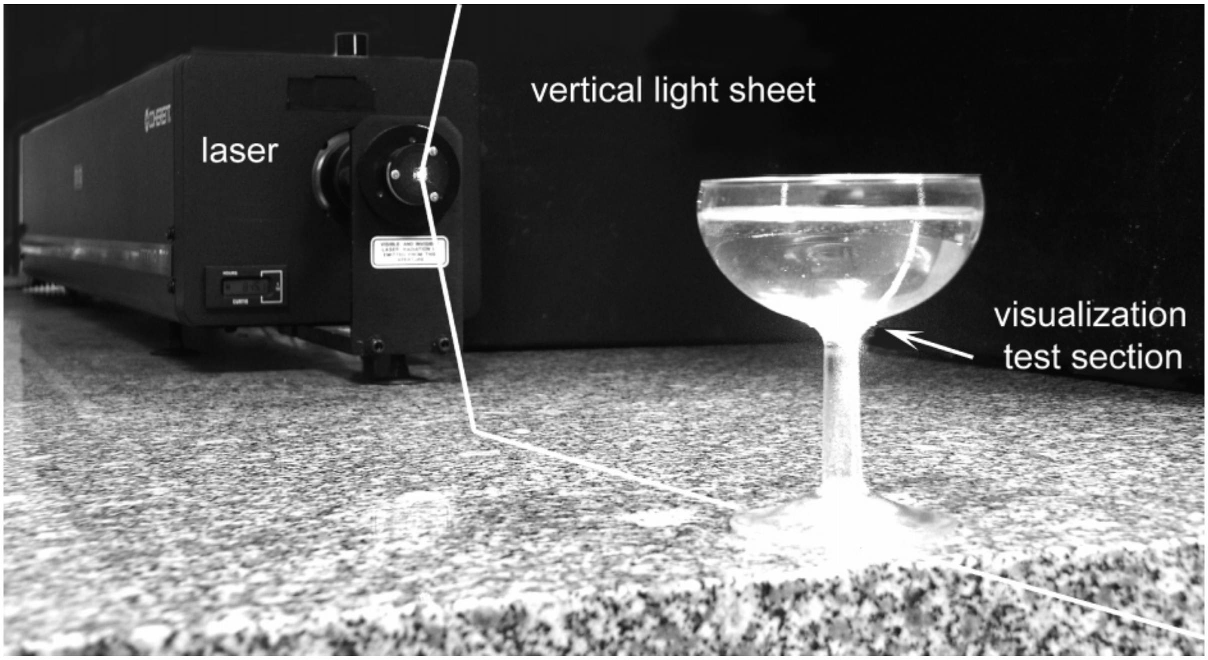

Figure 5. Optical bench used to visualize the flow pattern in champagne glasses.

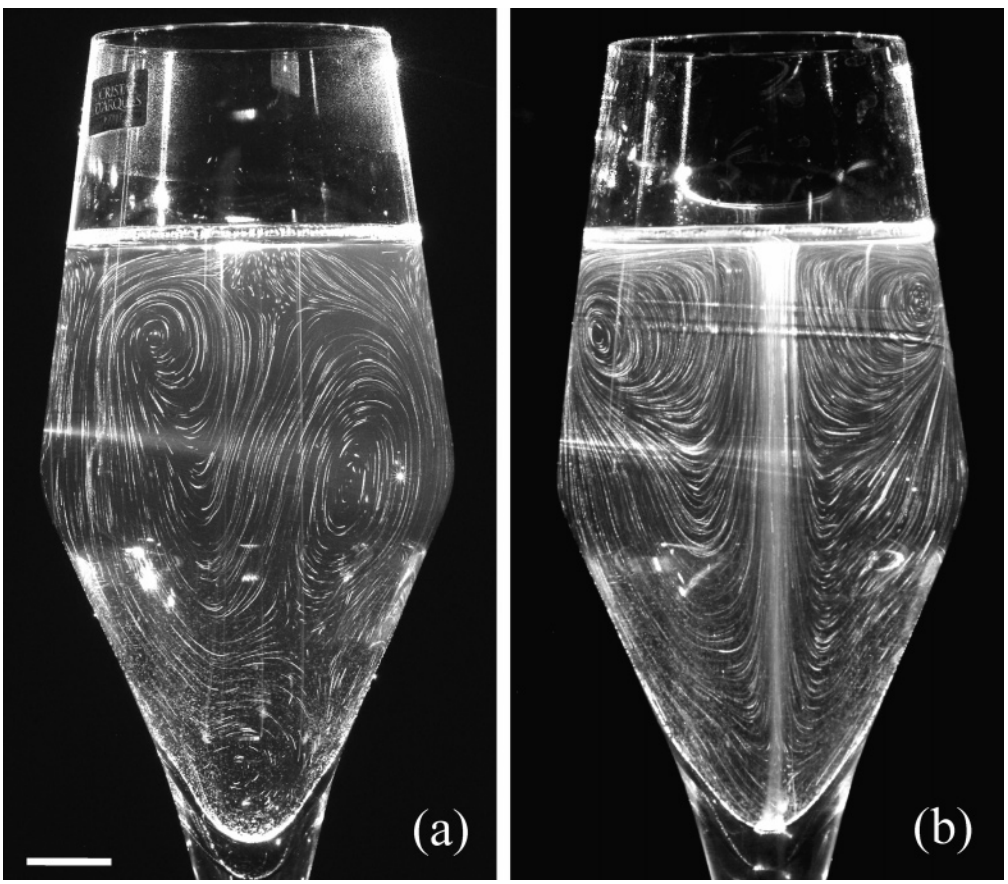

Figure 6. Visualization of the mixing flow patterns in the traditional flute without specific surface treatment showing natural effervescence (**a**) and in the traditional champagne flute engraved at its bottom (**b**). Bar = 1 cm.

steady flow regimes are identified for such a glass-shape. One regime clearly exhibits a 2-D axisymmetrical single swirling-ring (annulus) whose cross-section visualization reveals in **Figure 8** two counter-rotative vortices close to the glass axis. What strongly differs from the champagne flute is that this recirculating flow region does not occupy the whole volume in the glass. As a consequence, a singular steady flow regime is observed in the external periphery of the glass, which is also axisymmetric and characterized by a dead-zone of no motion. It means that, for a wide-brimmed glass, only about half of the liquid bulk participates in the champagne mixing process.

Beyond the visually appealing flow pattern phenomena revealed by this set of experiments, a question inevitably raises: what should be the enological consequences for the

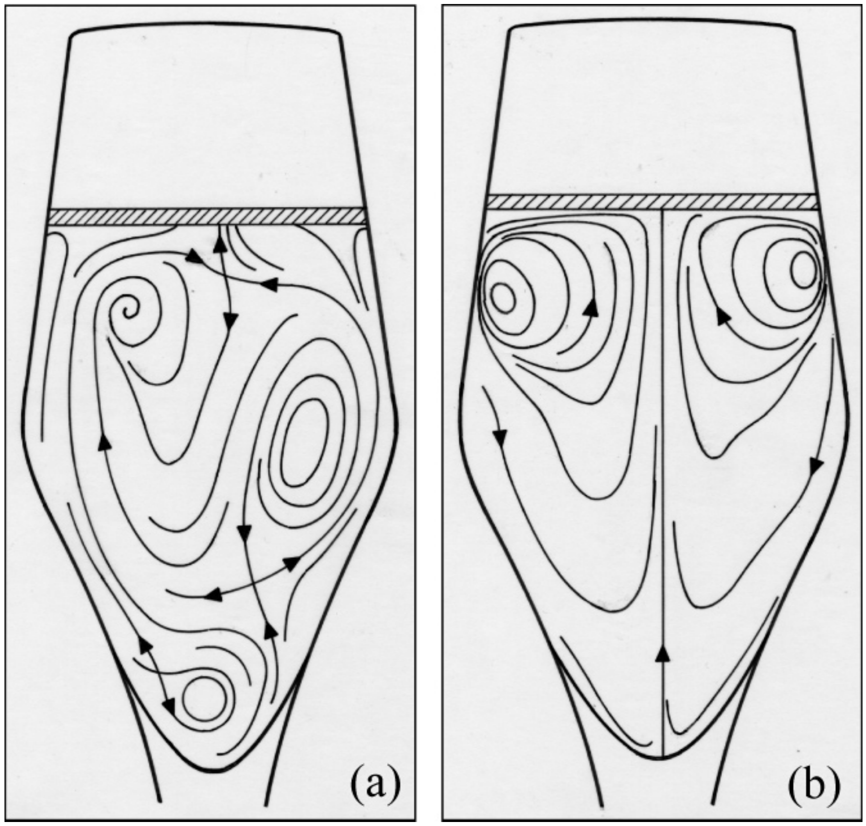

Figure 7. Streamline patterns in the traditional flute showing natural effervescence (**a**) and in the traditional champagne flute engraved at its bottom (**b**).

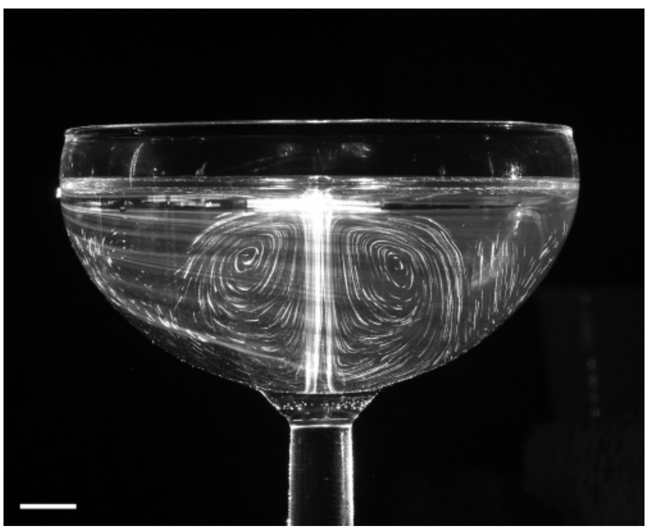

Figure 8. Flow pattern visualization inside the engraved champagne coupe. Bar = 1 cm.

consumer of such different champagne mixing processes depending on the glass-shape and engravement conditions? The aim of the two following paragraphs is to discuss the enological consequences of different champagne mixing processes during champagne tasting.

Impact on the Bubbles' Behavior at the Liquid Surface. During champagne tasting, consumers and judges often pay attention to the aspect of the foam ring at the edge of the flute or coupe, the so-called collerette or collar. To feed the collar, bubbles nucleated in the glass (by natural or artificial effervescence) must obviously reach the edge of the glass before bursting. Therefore, the formation of the collar is clearly ruled by a competition between the average bubble's lifespan at the liquid surface and the average time needed for a freshly emerged bubble to reach the edge of the glass. Coupes and flutes engraved at their bottom's surface close to their axis of symmetry (as the ones used in the present work) feed the liquid surface with freshly nucleated bubbles emerging in the middle of the liquid interface. Once arrived at the liquid surface, bubbles naturally migrate following the 2-D flow streamlines of the interface. At the liquid surface, coupes and flutes engraved at their bottom's surface close to their axis of symmetry show 2-D flow streamlines isotropically migrating from the center of the surface area toward the edge of the glass (as shown in **Figures 7b** and **8**). Consequently, once they reach the liquid surface, bubbles freshly emerged from the liquid bulk experience isotropic radial migration from the center of the surface area toward the periphery of the glass. The extent of this radial migration area is closely related to the bubbles' lifespan. In case of a widened glass such a the traditional coupe, bubbles cannot reach the glass edges during their lifespan contrary to the case of narrow glasses such as the traditional champagne flute where the average time needed by a bubble to reach the glass edge is smaller than the average bubble's lifespan, as seen in **Figure 9**. During the long exposure time ($t = 2$ s) of our camera, the trajectory of bubbles in **Figure 9** (inside the liquid bulk and at the liquid surface) is materialized by white filaments due to their very high degree of reflectivity with regard to the laser sheet. Actually, in view of **Figure 9a**,**b**, it is clear that a causal relationship exists between the radial migration extent of the bubbles and the size of the vortical flow that is below. Actually, in case of the champagne flute, the convective cells below the liquid surface literally carry the emerged bubbles from the center of the surface area toward the glass edge. In case of the champagne coupe, the convective cells carry the bubbles over a distance of only about 1 cm. Then, to reach the glass edge, bubbles must travel the dead-zone in terms of flow

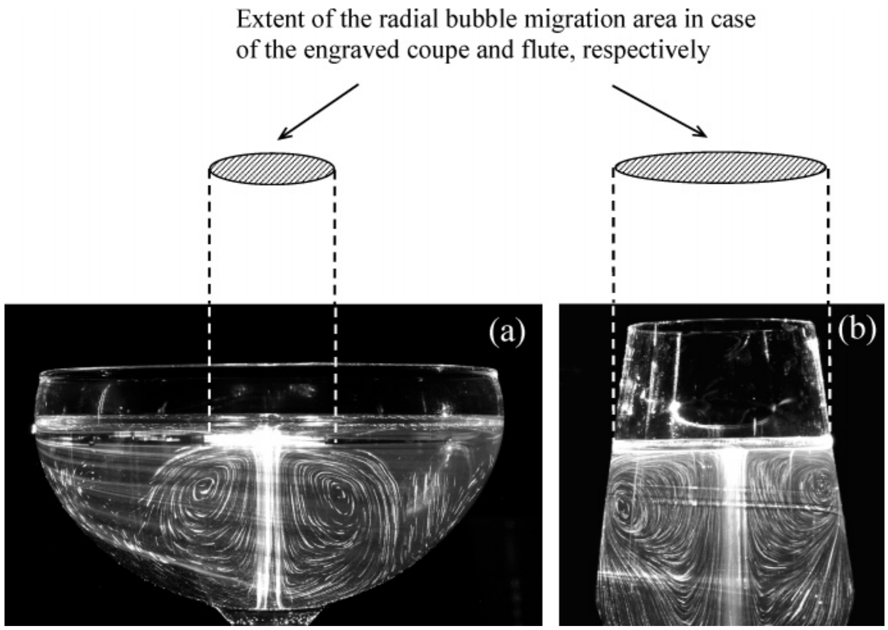

Figure 9. Extent of the radial bubble migration area at the liquid surface, in the case of the traditional engraved coupe (**a**) and flute (**b**), respectively.

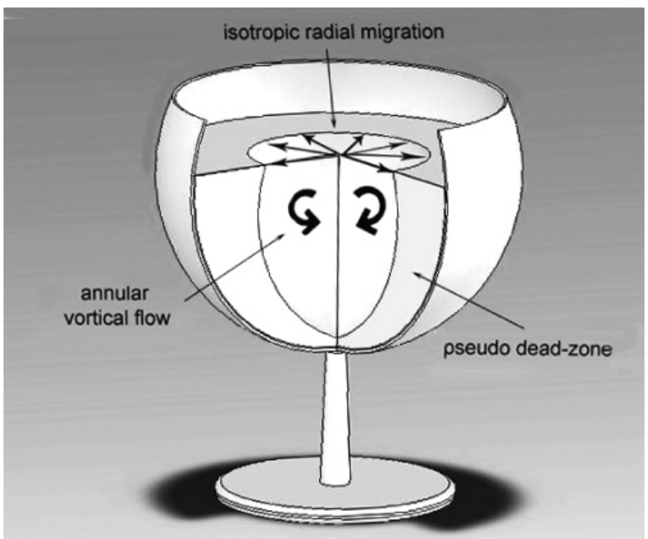

Figure 10. Scheme of the isotropic radial bubble migration process and identification of the two distinct zones in the bulk of the engraved champagne coupe.

streamlines. Most of them burst before reaching the edge of the glass to feed the collar (due to both a long distance to travel and unfavorable 2-D flow streamline conditions). A scheme is displayed in **Figure 10**, which illustrates the isotropic radial bubble migration process, as well as the two distinct zones in terms of flow motion in the bulk of the engraved champagne coupe.

Finally, both a short distance to travel for bubbles and favorable 2-D flow streamline conditions are the reasons why flutes are much more appropriate than coupes to observe a well-formed collar of bubbles very much sought after by champagne tasters.

Likely Impact on Flavor and Gas Release from the Liquid Surface. Generally speaking, gas discharge and flavor release from a liquid medium are ruled by the transfer of compounds from the liquid phase to the atmosphere through the liquid surface. The flux $\vec{J}_i$ of a given compound i through an interface is a combination of the diffusive and convective fluxes and is locally expressed by the first and second Fick's law as

$$\begin{cases} \vec{J}_i = c_i\vec{V} - D_i\vec{\nabla}c_i \\ \dfrac{\partial c_i}{\partial t} = \mathrm{div}(D_i\vec{\nabla}c_i) - \mathrm{div}(c_i\vec{V}) \end{cases} \qquad (1)$$

where $\vec{V}$, c_i, $\vec{\nabla}c_i$, and D_i are the velocity of the liquid flow, the bulk concentration of the given compound, the concentration gradients of the given compound close to the interface, and the diffusion coefficent of the given compound in the champagne bulk, respectively.

It is clear from eq 1 that both gas discharge and flavor release are highly fluid velocity-dependent through the parameter $\vec{V}$. The higher the fluid velocity V close to the liquid surface is, the higher the flux of a given compound through the liquid surface will be. The carbon dioxide release from the surface of champagne is therefore directly under the influence of the recirculating flow regions below the liquid surface. Despite the fact that this degassing process is totally invisible to the naked eye, it is much more important in quantity than the visible gas discharge directly linked with the release of carbon dioxide bubbles through effervescence as shown in a previous paper (*3*). Therefore, because the liquid bulk is much more vigorously mixed by the recirculating flow regions found in engraved glasses than in smooth glasses (as shown in **Figure 6**), the kinetics of carbon dioxide discharge and flavor release are strongly expected to be faster from champagne and sparkling wines poured into engraved flutes and coupes than from those poured into smooth flutes and coupes. From the consumer's point of view, because the release of a sudden and abundant quantity of CO_2 above the liquid surface is known to irritate the nose during the evaluation of aromas, glassmakers should pay better attention to quantify this gas discharge from engraved glasses. Experiments are to be conducted to evaluate the impact of various glass shape and engravement conditions on champagne and sparkling wines tastings.

Otherwise, as concerns champagne and sparkling wines poured into an engraved coupe, in view of the **Figure 8**, it is worth noting that the gas discharge and flavor release should not be homogeneous at all through the whole liquid surface.

Actually, the liquid surface above the dead-zone area characterized by a no-motion liquid bulk zone is expected to release fragrances and carbon dioxide molecules at a much lower rate than the liquid surface above the recirculating flow region around the axis of symmetry.

In conclusion, for the very first time, a classical flow visualization technique was used to capture the fluid motion in traditional flutes and coupes poured with champagne. It was found that glasses engraved around their axis of symmetry produce a rising gas column along the vertical glass axis that induces, in turn, recirculating flow regions. In the case of the classical engraved champagne flute, the whole domain of the liquid phase is homogeneously mixed, whereas in the case of the engraved champagne coupe, the recirculating flow region does not occupy the whole volume in the glass. In the engraved coupe, a dead-zone of no motion was identified, which inhibits the formation of the collar at the glass edge. Because the kinetics of flavor and gas release also strongly depends on the velocity of the recirculating flows close to the interface, we therefore strongly believe that this paper brings objective elements and clues to better understand the role of glass-shape and engravement conditions on the olfactive behavior of champagne and sparkling wines in a glass. In the near future, we plan to link our observations with quantitative measurements of the release of volatile organic compounds and carbon dioxide from glasses showing various engravement and shape conditions, our final goal being to scientifically identify the best glass for the tasting of champagne and sparkling wines in terms of gas discharge and flavor release.

ACKNOWLEDGMENT

Thanks are due to ARC International for supplying us with various glasses and to Virginie Deram and Hélène Lecoq from CREATE for supplying us with scanning electron microscopic views of laser beam impacts.

LITERATURE CITED

(1) Liger-Belair, G. *Uncorked: The Science of Champagne*; Princeton University Press: Princeton: NJ, 2004.

(2) Liger-Belair, G.; Vignes-Adler, M.; Voisin, C.; Robillard, B.; Jeandet, P. Kinetics of gas discharging in a glass of champagne: The role of nucleation sites. *Langmuir* **2002**, *18*, 1294−1301.

(3) Liger-Belair, G. La physique des bulles de champagne. *Ann. Phys. Fr.* **2002**, *27*, 1−106.

(4) Voisin, C.; Jeandet, P.; Liger-Belair, G. On the 3-D reconstruction of Taylor-like bubbles trapped inside hollow cellulose fibers acting as bubble nucleation sites in supersaturated liquids. *Colloids Surf., A* **2005**, *263*, 303−314.

(5) 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.

(6) Liger-Belair, G.; Tufaile, A.; Jeandet, P.; Sartorelli, J.-C. Champagne experiences various rhythmical bubbling regimes in a flute. *J. Agric. Food. Chem.* **2006**, *54*, 6989−6994.

(7) Voisin, C. Quelques aspects de la nucléation des bulles dans une flûte et de leur ascension à petits nombres de Reynolds. Ph.D. Thesis, Université de Reims Champagne-Ardenne, Reims, France, 2005.

(8) 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.

(9) Ronteltap, A. D.; Hollemans, M.; Bisperink, C. G.; Prins, A. Beer foam physics. *Master Brew. Assoc. Am., Tech. Q.* **1991**, *28*, 25−32.

(10) Lynch, D. M.; Bamforth, C. W. Measurement and characterization of bubble nucleation in beer. *J. Food Sci.* **2002**, *67*, 2696−2701.

(11) Liger-Belair, G.; Lemaresquier, H.; Robillard, B.; Duteurtre, B.; Jeandet, P. The secrets of fizz in Champagne wines: A phenomenological study. *Am. J. Enol. Vitic.* **2001**, *52*, 88−92.

(12) Merzkirch, W. *Flow Visualization*, 2nd ed.; Academic Press Inc.: London, 1987.

(13) Fohanno, S.; Polidori, G. Effect of the gap size in the start-up free convective flow around a square prism near a wall. *Int. J. Heat Fluid Flow* **2005**, *26*, 25−33.

(14) Zare, R. N. Strange fizzical attraction. *J. Chem. Educ.* **2005**, *82*, 673−674.

(15) http://www.stanford.edu/group/Zarelab/guinness/index.html.

Received for review October 16, 2006. Revised manuscript received November 24, 2006. Accepted November 28, 2006. Thanks are due to the Europôl'Agro institute, to the Association Recherche Œnologie Champagne Université, and to ARC International for financial support.

JF062973+