

## Bubble dynamics created by plasma in heptane

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The growth and collapse phases of bubbles created by micro-gap discharges in heptane (up to 5kV – 100 A during less than 1  $\mu$ s) are studied. Ultrafast video acquisitions are performed to determine the evolution of the bubble shape versus time. This evolution is compared to Rayleigh-Plesset and Gilmore solutions of the bubble movement. In the present situation, good agreement between experimental and theoretical data can be achieved with the Gilmore model.

Bubble dynamics in liquids have been intensively investigated to better understand how this phenomenon influences applications where it is encountered. Namely, electro-discharge machining [1], sonoluminescence [2] or nano-material synthesis [3] are fields where the successive phases of growth and collapse of bubbles need to be understood and controlled.

Many models are available to describe the bubble dynamics, among which one finds the Rayleigh-Plesset model:

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{P_i}{\rho_0} \quad (1)$$

where  $\rho_0$  is the liquid density at room temperature.  $P_i$  is the dynamical pressure.  $R$  is the bubble radius,  $\dot{R}$  and  $\ddot{R}$  are the first and second time derivatives of  $R$ , *i.e.* the bubble wall velocity and acceleration.  $P_i$  may be written as:

$$P_i(t) = P_{gas}(R, t) - 4\mu\frac{\dot{R}}{R} - \frac{2\sigma}{R} - P_0 \quad (2)$$

$P_0$  is the pressure in the liquid far from the bubble wall, *i.e.* atmospheric pressure.  $-\frac{2\sigma}{R}$  and  $-4\mu\frac{\dot{R}}{R}$  describe the effect of the surface tension  $\sigma$  and the fluid viscosity  $\mu$  respectively. The gas pressure in the bubble is given by  $P_{gas}$ . This term depends on  $R$  and  $t$ . To account for this dependance, one needs to resort to an equation of state for the bubble, which is generally a  $\gamma$ law.

$$P_{gas}(R, t) = \left( P_0 + \frac{2\sigma}{R_0} \right) \left( \frac{R_0}{R} \right)^{3\gamma} \quad (3)$$

In this study,  $\gamma=5/3$ . On the other hand, the Gilmore equation includes a thermodynamic part in the behavior of the liquid and this approach can be used at high Mach numbers. One has:

$$\left( 1 - \frac{\dot{R}}{C} \right) \frac{R}{C} \frac{dH}{dt} + \left( 1 + \frac{\dot{R}}{C} \right) H - R\ddot{R} \left( 1 - \frac{\dot{R}}{C} \right) - \frac{3}{2} \left( 1 - \frac{\dot{R}}{3C} \right) \dot{R}^2 = 0 \quad (4)$$

Here,  $H$  is the enthalpy difference between the liquid at pressure  $P$  and the liquid at pressure  $P_0$ .  $C$  is the sonic velocity. The Gilmore's model must include an equation of state for heptane giving  $H$  and  $C$  as a function of  $P$ . The properties of the liquid phase become pressure-dependent. We took values available in reference works [4, 5]. For heptane,  $\sigma = 0.019816$  N m<sup>-1</sup> and  $\mu = 0.0003805$  Pa s.

In the present study, we used liquid heptane. Discharges are made in a tip-to-plane configuration, between a platinum wire embedded in a silica capillary to set accurately its area. The counter-electrode is made of aluminium. The growth and collapse phases of bubbles created in a 50- $\mu$ m gap configuration by DC discharges (up to 5kV – 100 A during less than 1  $\mu$ s) are studied. Ultrafast video acquisitions (525 kHz, *i.e.* recording time 1.905  $\mu$ s) are performed to determine the evolution of the

bubble shape versus time. The shape of the bubble is shown in figure 1. It is not spherical. An equivalent spherical radius  $R$  is then determined from the volume of revolution given by rotation on its symmetry axis of the surface captured on the photograph. Experimental and modelling results are given in figure 1. A fairly good agreement is obtained with the Gilmore model. The main parameters needed to fit  $R(t=0) = 50 \mu\text{m}$ ,  $R_0 = 137 \mu\text{m}$ ,  $\dot{R}(t=0) = 250 \text{ m s}^{-1}$ . For the Rayleigh-Plesset model, the temporal characteristics of the bubble oscillations and their attenuation can be reproduced accurately, assuming  $R(t=0) = 20 \mu\text{m}$ ,  $R_0 = 60 \mu\text{m}$ ,  $\dot{R}(t=0) = 1500 \text{ m s}^{-1}$ . However, the amplitude of oscillations is overestimated by  $\sim 15\%$ . In this case, the density being treated as constant, the bubble behaves as it was lighter. Then, the initial impulse imparted to the bubble wall and its reference radius must decrease to keep the same temporal characteristics.

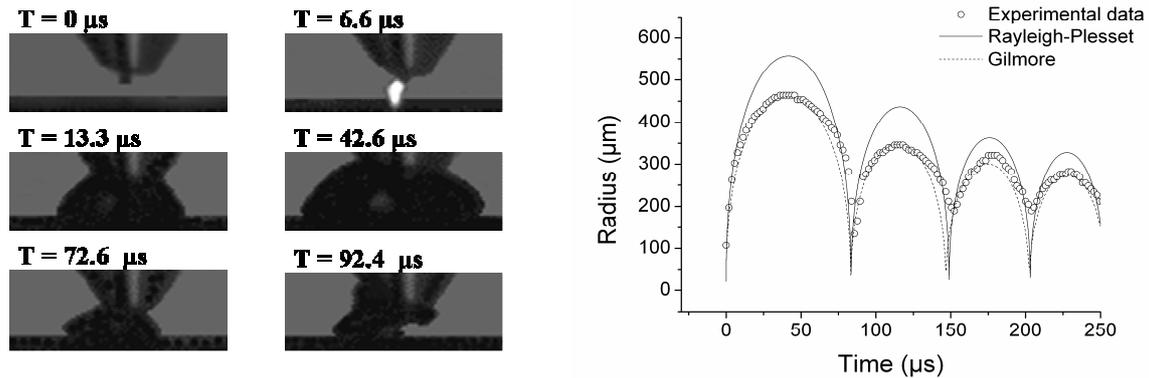


Fig. 1: Left: Selected pictures of ultrafast acquisitions of bubble dynamics. Right: comparison between experimental data and modelling results

The determination of the initial pressure from each model gives here comparable results:  $P(t=0) = 1.53 \times 10^7 \text{ Pa}$  for Gilmore and  $P(t=0) = 2.28 \times 10^7 \text{ Pa}$  for Rayleigh-Plesset. We determine by emission spectroscopy that the plasma temperature is about 5000 K. In these conditions, nanodiamond synthesis is possible, as shown by [6]. To go a step further, we would need to get information between  $t = 0$  and  $t = 1.905 \mu\text{s}$  to access accurately to the initial velocity of the bubble wall. Electrical measurements show that the plasma phase lasts for about 500 ns. However, it is well known that the identification of the bubble just after the plasma phase is difficult. It turns out that if the bubble dynamics can be described accurately, the very first instants raise specific difficulties that would need to be overcome to better understand the transition from the plasma to the bubble.

## References

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