

## Explosions of oxygen bubbles in cyclohexane

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### Abstract

Experimental investigations of the explosive behavior of pure oxygen bubbles in cyclohexane have been performed. For the investigations an autoclave containing a bubble generator was used and optical and pressure measurements were carried out. A shock wave was used to ignite the bubbles. The present work is the first where the explosion behavior of the bubble was investigated not only in the first oscillations after the incident shock wave passage, but for many oscillations after it. From these investigations two bubble ignition types are registered. One type of explosion is the well-known bubble explosion occurring in the first bubble oscillation after the shock wave impact. The other type was unknown up to now. It takes place after many bubble oscillations and has a significantly longer delay time before ignition.

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### 1. Introduction

Systems consisting of a fluid as the matrix and a gas as a dispersed phase have for many decades attracted investigators because of their special properties [1]. Some studies have been made investigating the explosive behavior of a bubbly medium under shock wave [2–4]. Bubble ignition was found to be possible in the following systems, referred as systems I and II. In system I, the liquid is inert and the gas phase of the bubbles explosive, e.g. water with bubbles of hydrogen–oxygen mixtures. In system II, a fuel and its oxidizer are in separate phases, e.g. a liquid organic solvent containing oxygen bubbles.

A similar ignition behavior in both systems was observed. When a bubble explosion was obtained in system I, the bubble ignition took place in the first oscillation following the incident shock wave impact [2–6]. The bubble ignition in system II, was found to present a similar behavior as in system I [7–9]. The main difference of the bubble ignition between these two systems is that in system II the presence of mass injection, evaporation and mixing processes in the gas phase of the bubble is necessary. It was also found [8,10,11] that in bubble columns of systems I and II the formation of a self-sustaining bubble detonation wave is possible.

It is interesting to note here that until today only the possibility of a bubble explosion after a relatively short-time, i.e. only in the first bubble oscillations after the passage of the incident shock wave, has been investigated. This has left open the question of the bubble behavior in later stages after the shock wave loading. This question is interesting especially in system II, where the interaction between the liquid and the gas phase of the bubble plays a critical role in the explosion behavior of the latter. A further interest on this point rises from the fact that hydrocarbons can give explosion regimes with a variety of explosion mechanisms and explosion parameters [12]. These mechanisms may vary from high temperature ignition to low temperature two stages ignition. Additionally, cool flames can be involved.

The investigations in system II and especially those involving an organic solvent and oxygen bubbles are very rare [3,9]. Furthermore, there is only one known investigation [8] referring to the intermediate system: liquid fuel with oxidizer bubbles where the vapor pressure of the liquid, initial pressure and temperature of the system allow the bubble to contain an explosive gas mixture before the shock wave impact. An example of such an intermediate system is liquid cyclohexane containing initially pure oxygen bubbles, under normal conditions.

The lack of experimental work with system II in general, and with these intermediate systems in particular, is very surprising considering the fact that such chemical systems

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are widely used in the chemical industry. Such media are required to conduct processes, such as liquid-phase hydrocarbon oxidation. An example is the oxidation of cyclohexane to cyclohexanone and cyclohexanol, which is used as raw material for the production of nylon.

The tendency of the industry towards more extreme conditions (e.g. use of pure oxygen instead of air in oxidation processes) is caused by the need for cost and quality optimization of the production [13,14]. An indication for the existence of this tendency can be found also in the general direction of the patents from the industry. For example, the patents for the oxidation of cyclohexane were referring to simple air bubbles 30 years ago [15], while new patents during the last years already involve bubbles containing 90% oxygen [16].

For these reasons, we performed our investigations for the system liquid cyclohexane containing pure oxygen bubbles under normal conditions, i.e. an intermediate system II. The present work is focused on the single bubble explosion behavior. This was done mainly for two reasons. First, because it is a necessary step in order to solve the problem of the bubble detonation wave initiation and propagation. Second, because the ignition and subsequent collapse of even single bubbles may lead to industrial chambers wall erosion as experiments have shown [17,18]. Furthermore, the present work is the first where the explosion behavior of the bubble was investigated not only in the first oscillations after an incident shock wave passage, but for many oscillations after it.

Two bubble ignition types are registered. Apart from the ignition behavior known in systems I and II, also a new ignition type was observed that takes place after many bubble oscillations and has a significantly longer delay time before ignition. Much more complex phenomena seem to dominate this new type of ignition.

## 2. Experimental

A scheme of the autoclave is shown in Fig. 1. The autoclave is designed pressure resistant for up to 1100 bar at 200 °C. The inside of the autoclave is a vertical cylindrical tube of 100 mm inner diameter and 1070 mm length. The bubble generator is installed in the bottom of it, and consists of an orifice with two side openings of 0.35 mm diameter each. The autoclave contains four holes of 100 mm diameter, in two of which the windows for the optical measurements are installed. In the other two holes, the adapters for the pressure measurements are installed.

The experimental procedure was the following. First the appropriate volume of liquid is imported into the autoclave. Then an explosive acetylene–oxygen gas mixture is fed in from the upper gas inlet. Bubbles are created by injecting gas (oxidizer or inert) into the liquid phase through the bubble generator on the bottom. A shock wave in the liquid is generated by a gas detonation of the explosive mixture

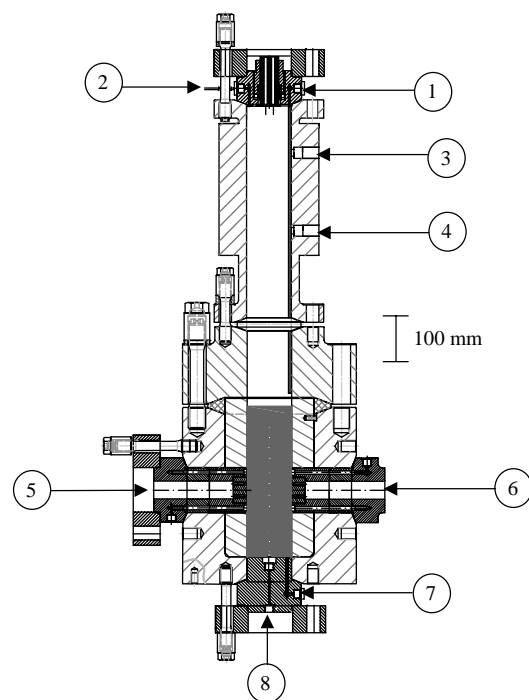


Fig. 1. Scheme of the autoclave: (1) gas inlet; (2) gas outlet; (3–6) pressure sensor positions; (7) liquid outlet; (8) gas inlet for the bubbles.

above the liquid. The gas detonation is ignited by an exploding wire in the top flange of the autoclave. The gas and liquid outlets are used for the evacuation of the autoclave at the end of the experiment.

For the pressure measurements, piezoelectric pressure transducers (Kisler 601H) in combination with a multi-channel transient recorder operating at a sample rate of 1 MHz were used. The detonation pressure in the gas phase is measured at the positions 3 and 4 (see Fig. 1). The resulting shock wave pressure in the liquid is monitored at two positions on each of the two adapters. The vertical distance between these two positions is 66.4 mm.

The interaction of the bubbly liquid with the incident shock wave was observed with the help of high-speed digital framing photography using either a diffuse or parallel light as external light source.

In all the experiments presented here, the liquid phase amounted to 4750 ml pure cyclohexane. Its surface was situated 350 mm from the bottom of the autoclave, reaching the level shown in Fig. 1. The mole fraction of acetylene and oxygen in the explosive mixture above the liquid phase were 0.25 and 0.75, respectively. The initial pressure of the mixture was 1 bar. The bubbles in our system were typically of non spherical shape. The equivalent bubble diameter was typically  $3.0 \text{ mm} \pm 0.5 \text{ mm}$ . The equivalent diameter was defined by:  $d_e = (ab^2)^{1/3}$ , where  $a$  and  $b$  are the minor and major axes of the bubble, respectively. All experiments were performed at room temperature (20–25 °C).

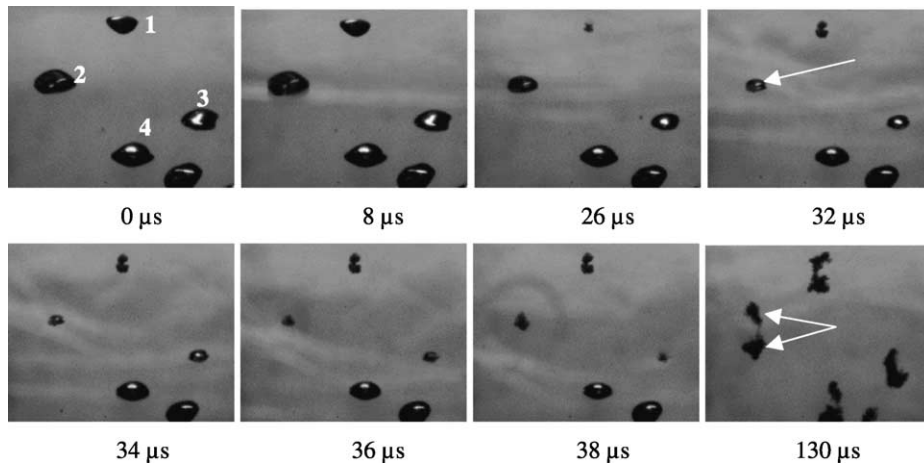


Fig. 2. Nitrogen bubbles under shock wave. Just before the entrance of the incident shock wave into the observation window ( $0 \mu\text{s}$ ). The incident shock wave is in the middle of the observation window ( $8 \mu\text{s}$ ). Maximum levels of compression for first bubble ( $26 \mu\text{s}$ ). Strong jet formation in the second bubble ( $32 \mu\text{s}$ ). The second bubble is broken into two parts. Recording rate: 500,000 fps. Exposure time per frame:  $2 \mu\text{s}$ .

### 3. Results

Experiments with  $\text{O}_2$  bubbles under the above conditions were carried out. For comparison, we performed experiments with  $\text{N}_2$  bubbles under the same conditions also. The oxygen bubbles showed two different explosion behaviors, a distinctive macroscopic difference between them being the different delay time before ignition.

#### 3.1. Nitrogen bubbles under shock wave

For illustration of the shock-induced bubble dynamics, the frames from an experiment with nitrogen bubbles, recorded

at a speed of 500,000 fps are presented in Fig. 2. The following stages of shock-induced bubble behaviour were observed. After some time delay, the bubble starts to shrink. During the compression a jet forms, which penetrates the bubble (e.g. see Fig. 2,  $32 \mu\text{s}$ , bubble no. 2). The main reason for this jet formation is the inhomogeneous pressure field in the liquid near the bubble, as well as surface instabilities and the non symmetrical bubble form. After the jet formation, the bubble compression continues and two different behaviors were observed. Either the bubble breaks up into two parts (e.g. see Fig. 2,  $130 \mu\text{s}$ , bubble no. 2), or the bubble remains unbroken and reaches a minimum diameter which is followed by an expansion phase (e.g. see Fig. 2, frames

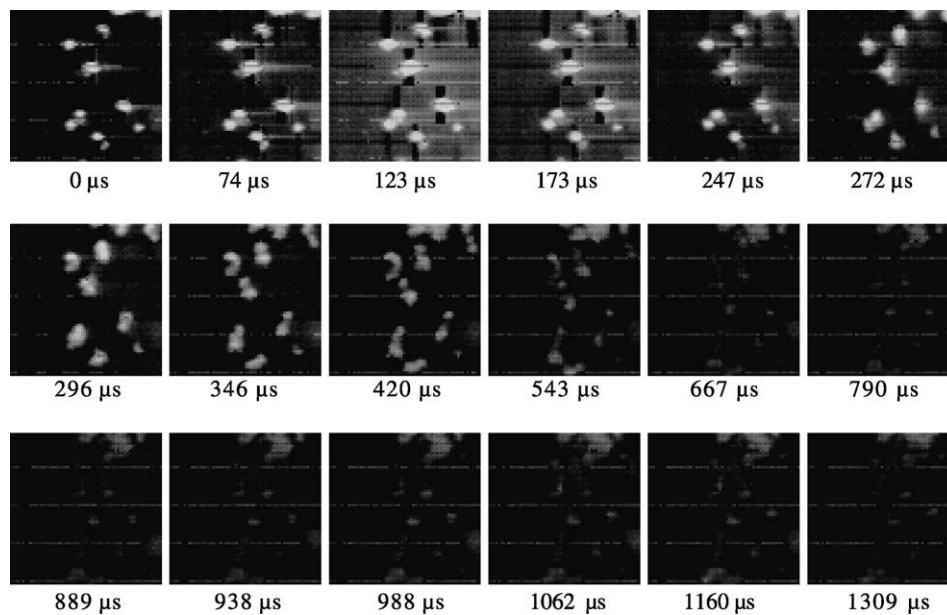


Fig. 3. Nitrogen bubbles under shock wave: detonation wave impact on the surface ( $123 \mu\text{s}$ ); shock wave passage through the bubbles ( $272 \mu\text{s}$ ). Recording rate: 40,500 fps. Exposure time per frame:  $24.69 \mu\text{s}$ .

between 26 and 130  $\mu\text{s}$ , bubble no. 3). The initial equivalent diameters of the bubbles shown in these frames, were in the range 3.0–3.6 mm. The peak pressure of the incident wave was 54 bar.

The camera used to record the frames presented in Fig. 2 offers a high time resolution but a limited total number of recorded frames. Therefore, a second camera was used, which offers a much higher total number of frames and recording time. The time resolution and optical quality of these frames is not as high as of the first camera. Nevertheless, this camera was necessary to be used in order to observe the explosion behavior of the bubbly medium for longer times with an acceptable time resolution (i.e. 40,500 fps).

For comparison with the oxygen experiments recorded by this camera, an experiment with nitrogen bubbles is pre-

sented in Fig. 3. In this experiment, the incident shock wave in the liquid had a magnitude of 61 bar. The pressure signals of the detonation wave in the gas phase and the shock wave in the liquid phase are the signals A in Fig. 4. The data shown in Fig. 4 were recorded with a time resolution of 1  $\mu\text{s}$  and smoothed with a running average of 20 points. The smoothing was necessary for reducing the influence of the natural oscillations of the pressure transducers. Averaging over 20 points corresponds to exactly three oscillation periods of the transducers.

In the experiments with the nitrogen bubbles, the following were observed. During the propagation of the detonation wave in the gas phase towards the surface of the liquid, both the bubbles and the liquid are becoming brighter. This effect reaches its maximum as the detonation wave reaches the

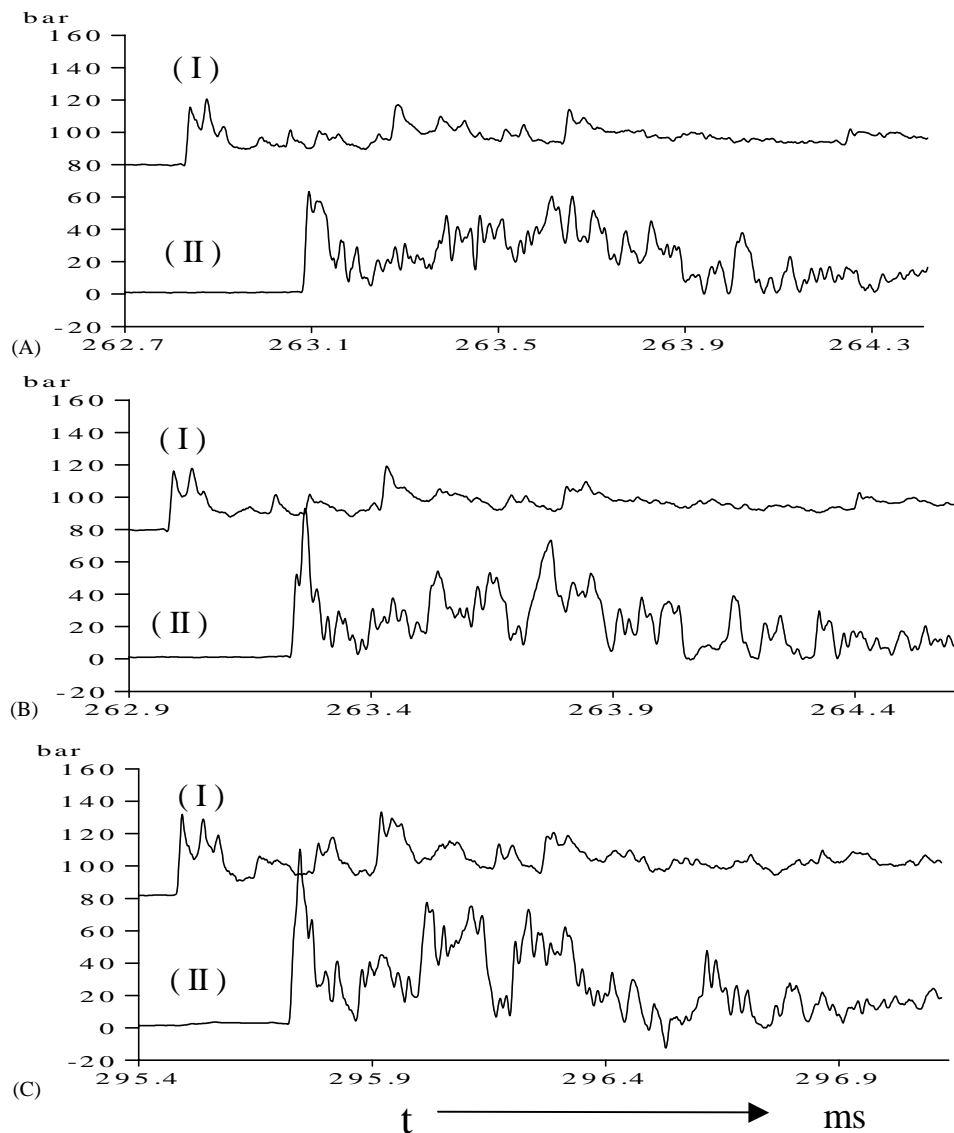


Fig. 4. Pressure signals of the experiment showing (A) nitrogen bubbles, (B) oxygen bubbles with short ignition delay and (C) oxygen bubbles with long ignition delay. The pressure of curves (I) correspond to the gas phase and was measured at position 4 in Fig. 1. The pressure of curves (II) correspond to the liquid phase and was measured at position 6 in Fig. 1. A pressure of 80 bar is artificially added to all pressure signals in the gas phase, to make them distinguish from the pressure signals in the liquid phase.

surface of the liquid (Fig. 3, 123  $\mu\text{s}$ ) and is reduced rapidly thereafter, reaching an almost constant level. Bubble and liquid illumination, caused by the light of the detonation wave propagating in the gas phase, was a common effect of all our experiments with nitrogen and with oxygen bubbles.

As shown in Fig. 2, the shock wave impact on the bubble causes a violent change of the bubble's size and form. The time delay after the shock wave impact for this change to be evident, is estimated to be generally about 10–15  $\mu\text{s}$  in our system (see Section 4). Each frame in Fig. 3 corresponds to about 25  $\mu\text{s}$  recording time. Therefore, the frame during which the passage of the shock wave through the bubbles took place, is the first one in which the bubbles' form appear to be changed. Having identified the frame during which the detonation wave from the gas phase hits the liquid surface, the approximate time between these two events can be calculated. This time corresponds to the time calculated directly from the shock wave propagation velocity in the liquid and the distance between the surface of the liquid and the center of observation.

### 3.2. Oxygen bubble ignition during the first shock-induced oscillation

We observed two different ignition delays ( $\tau_{\text{ign}}$ ) for the explosion of the oxygen bubbles under the same experimental conditions. In Fig. 5, an example of an experiment with the first type of bubble explosion, i.e. with a short ignition delay, can be seen. The incident shock wave in the liquid had a magnitude of 91 bar. This shock wave passed through the bubbles, during the recording of the frame at 222  $\mu\text{s}$ . The pressure signals of the detonation wave in the gas phase

and the shock wave in the liquid phase are displayed in Fig. 4.

The light effect from the detonation wave in the gas phase reached its maximum during the frame at 123  $\mu\text{s}$  (Fig. 5). After the passage of the shock wave (frame at 222  $\mu\text{s}$ , Fig. 5) a different bubble behavior as compared with the nitrogen bubbles was observed. The bubbles did not break into parts, but expanded and the light intensity related to the surrounding liquid was increased. These indicate the existence of a bubble explosion, shortly after the passage of the shock wave. Since the light emission from the gas phase above the liquid is still influencing considerably the optical measurements during the time period when this early bubble explosion takes place, one of the experiments with another highspeed digital camera is presented to clearly demonstrate this type of bubble explosion. The advantage of this camera is the higher quality of the frames produced, as can be seen. Due to the limited number of frames it can produce per experiment, it was not possible to record both bubble explosion behavior with this camera.

Frames of this experiment can be seen in Fig. 6. The incident shock wave in the liquid enters the observation window at 0  $\mu\text{s}$ . Its magnitude was 56 bar. The sequence of the frames shows that the passage of the shock wave through a bubble, makes the bubble first to shrink. During this compression phase the bubble explodes, which results in a sequence of single bubble explosions after the passage of the shock wave. This kind of early explosion had in our experiments a typical duration of light emission of a few microseconds and an ignition delay typically in the range of  $\tau_{\text{ign}} = 18\text{--}24 \mu\text{s}$ . This short delay before ignition and duration of light emission is reported also in other systems (e.g.

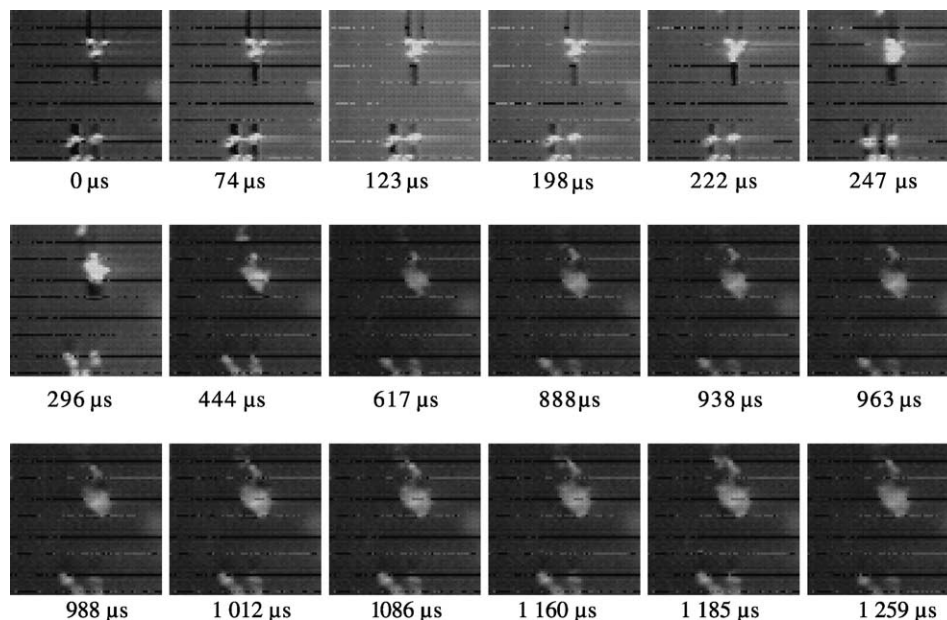


Fig. 5. Oxygen bubbles under shock wave: detonation wave impact on the surface(123  $\mu\text{s}$ ); shock wave passage through the bubbles (222  $\mu\text{s}$ ); bubble ignition (247  $\mu\text{s}$ ). Recording rate: 40,500 fps. Exposure time per frame: 24.69  $\mu\text{s}$ .



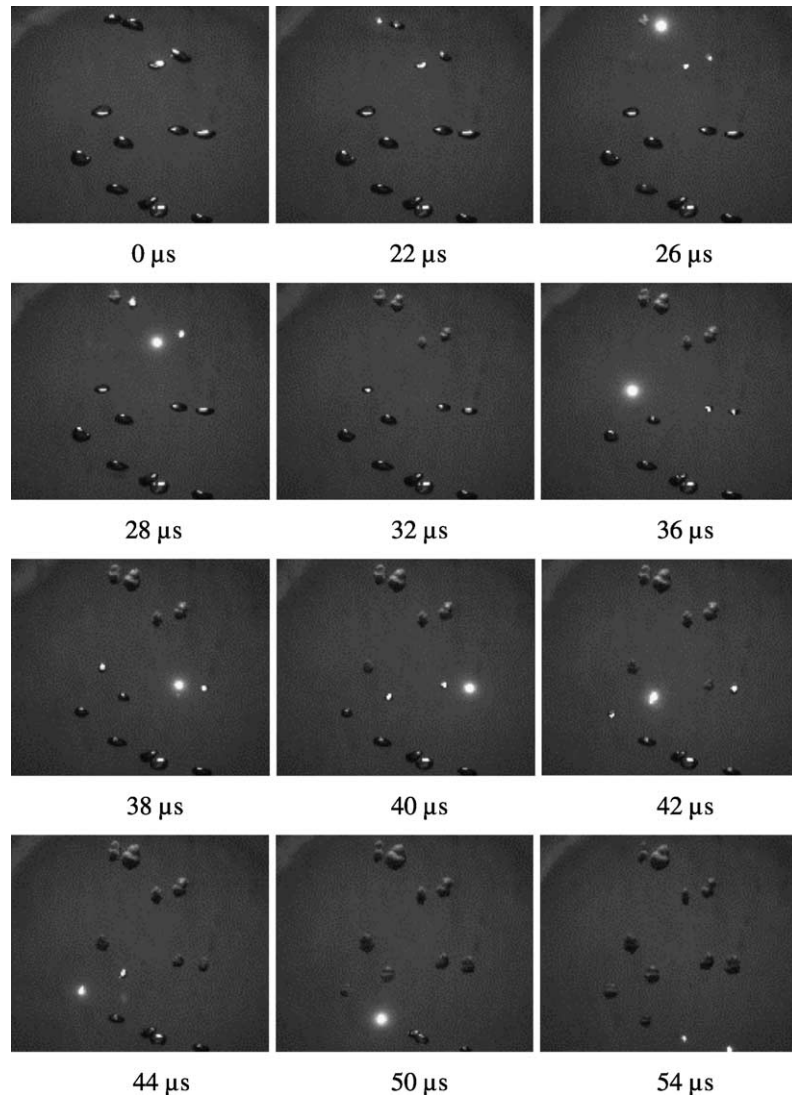


Fig. 6. Oxygen bubbles under shock wave. The incident shock wave is inside the observation window during the time 0–32  $\mu\text{s}$ . In the frames recorded during the time 26–50  $\mu\text{s}$ . Sequential bubble ignitions. Illumination of light from the gas phase above the liquid can be seen on the bubbles during the time before their explosion. Recording rate: 500,000 fps. Exposure time per frame: 2  $\mu\text{s}$ .

[4,6,7,19]). Not in all experiments we observed bubble ignition during the first compression phase after the incident shock wave impact. A second bubble explosion behavior, that is described in the following paragraph, was occasionally also observed. This kind of explosion behavior is not described in the literature up to now.

### 3.3. Oxygen bubble ignition after several oscillations

In Fig. 7 one can see a typical example of an experiment with the second type of bubble explosion, i.e. with a relatively long ignition delay. The incident shock wave had a magnitude of 105 bar. This shock wave passed through the bubbles, during the recording of the frame at 222  $\mu\text{s}$ . The pressure signals of the detonation wave in the gas phase and the shock wave in the liquid phase are the signals C in Fig. 4.

The light effect from the detonation wave in the gas phase reaches its maximum at the frame taken at 123  $\mu\text{s}$  (Fig. 7) (so during this frame the detonation wave reached the surface of the liquid). This optical effect is reduced thereafter reaching an almost constant level after the frame at 568  $\mu\text{s}$ . An unexpected expansion phase of the bubbles started at 617  $\mu\text{s}$  after the shock wave impact. This expansion is evident in Fig. 7 (938–1111  $\mu\text{s}$ ) and Fig. 8. An explanation of this expansion phase can not be cavitation effects, as a strong rarefaction wave that would create such effects was not observed during that time, (see curve C in Fig. 4). Therefore, it can be reasonably assumed that a bubble explosion takes place. It is interesting to note that as a result of the passage of the incident shock wave, bubble breakage proceeded this new type of bubble ignition.

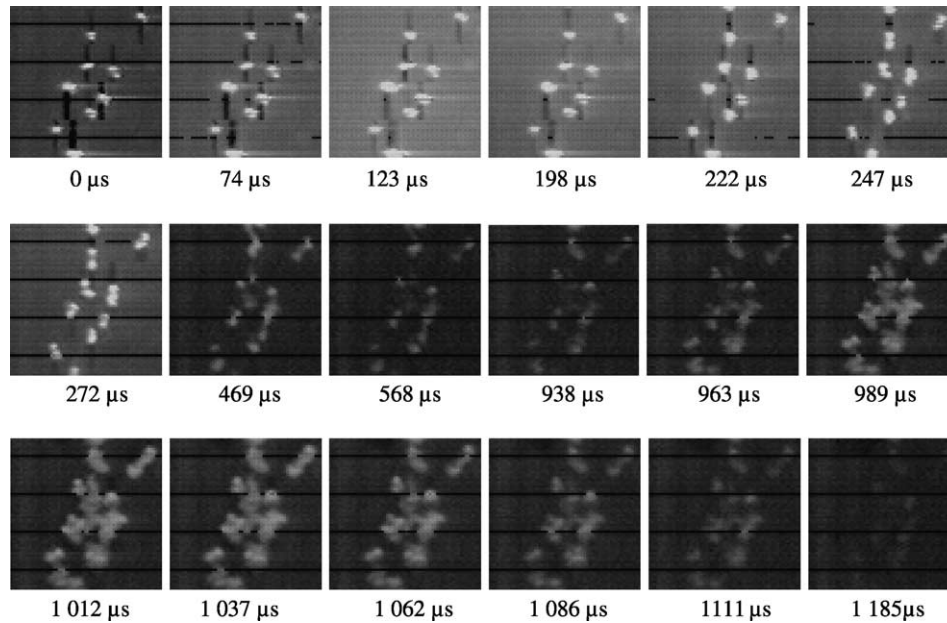


Fig. 7. Oxygen bubbles under shock wave: detonation wave impact on the surface (123  $\mu$ s); shock wave passage through the bubbles (222  $\mu$ s); bubble ignition (963  $\mu$ s). Recording rate: 40,500 fps. Exposure time per frame: 24.69  $\mu$ s.

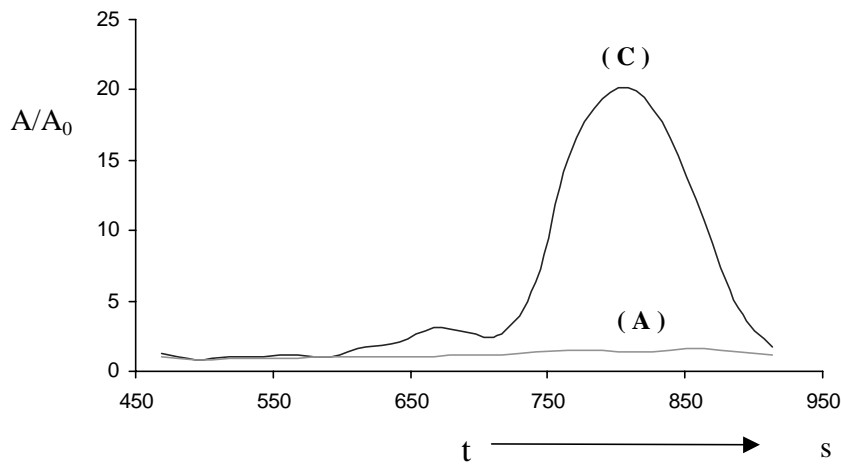


Fig. 8. Diagram of total bubble area divided with initial bubble area, as a function of time after the shock wave impact for (A) the nitrogen bubbles and (C) the oxygen bubbles with long delay before ignition. The curves (A) and (C) correspond to the signals (A) and (C) of Fig. 4, respectively.

In Fig. 8, the change of total visible bubble area as a function of the time from the shock wave impact is presented. This curve gives a qualitative information on the mean bubble diameter change as a function of the time. For comparison, the corresponding curve from the experiment with nitrogen bubbles is also included in the diagram. It is interesting to note the two peaks that the curve for the oxygen bubbles presents. This is a characteristic feature that we observed always in this type of bubble explosion. The bubble explosion starts at the beginning of the first peak, which is typically more than 600  $\mu$ s after the shock wave impact.

## 4. Discussion

### 4.1. Pressure signals

It was not possible to clearly identify either type of bubble explosion inside our pressure measurements. The reason of this must lay on the fact that the pressure waves from the single bubble explosion had to travel a relatively long distance (at least 5 cm) to meet the pressure sensors, making the damping effect important. A further difficulty originates from the fact that the structure of the pressure in the liquid has a very intense oscillatory structure and that stochastic

processes can have a strong influence in its properties too.

The propagation velocity of the shock wave inside the bubbly medium had always a value close to the sound speed of 1280 m/s [20] in the liquid. The measured spread of the shock wave propagation velocity between the experiments can be explained by the variation of the parameters that have an influence on it (e.g. bubble concentration in the liquid, magnitude of the detonation wave from the gas phase). After the passage of the incident shock wave, the pressure inside the liquid tends to oscillate around—and finally reach—a value, which is the pressure of the gas phase after the detonation. In our experiments, this value was about 10–20 bar at about 1 ms after the incident shock wave passage (see Fig. 4). The oscillatory structure of the wave inside the liquid phase is a known characteristic feature of a shock wave propagation inside a bubbly medium [21–24].

#### 4.2. Diffusion process in the bubble, before the shock wave impact

We fed in pure oxygen or nitrogen for the gas phase of the bubbles. During the rising of the bubble in the liquid cyclohexane, vapors of the shock wave are expected to have enriched the bubble's gas phase. Under the assumptions that: (i) only the molecular diffusion is responsible for this enrichment; (ii) the bubble is a sphere with constant radius  $\alpha$ ; (iii) interfacial equilibrium holds at  $r = \alpha$ ; (iv) the diffusion coefficient  $D$ , and the equilibrium concentration of cyclohexane in the bubble,  $C_{\text{eq}}$ , are constant; (v) the initial concentration of cyclohexane in the sphere is zero; and (vi) radial diffusion process takes place, the second Fick's law for diffusion applies. The solution for this equation, for spherical coordinates and under the above assumptions is given in [25]:

$$C(r, t) = C_{\text{eq}} + \frac{2a C_{\text{eq}}}{\pi r} \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \sin \frac{n \pi r}{\alpha} e^{(-D n^2 \pi^2 t)/\alpha^2} \quad (1)$$

where  $C(r, t)$  is the concentration of cyclohexane at the position  $r$  inside the bubble and at the time point  $t$ . From (Eq. (1)) it follows that the average concentration of cyclohexane in the bubble is:

$$C_{\text{av}} = C_{\text{eq}} - \frac{6C_{\text{eq}}}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{(-D n^2 \pi^2 t)/\alpha^2} \quad (2)$$

This equation can also be written as:

$$\frac{C_{\text{av}}}{C_{\text{eq}}} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-n^2(t/\tau)}, \quad (3)$$

where

$$\tau = \frac{\alpha^2}{\pi^2 D} \quad (4)$$

is the characteristic time of the process.

Table 1

$t/\tau$	$C_{\text{av}}/C_{\text{eq}}$
0.001	0.045
0.010	0.105
0.100	0.310
0.200	0.421
0.500	0.610
1.000	0.774
2.000	0.918
3.000	0.970
4.000	0.989
5.000	0.996
6.000	0.998
7.000	0.999
8.000	1.000

In Table 1, calculated values for  $(C_{\text{av}}/C_{\text{eq}})$  for some typical values of  $t/\tau$  are presented.

It can be seen that after the characteristic time  $\tau = \alpha^2/(\pi^2 D)$ , the saturation level of the gas bubble in cyclohexane vapors is about 77%. After  $3\tau$  the bubble has reached 97% of the saturation level.

For a binary gas mixture, and for temperatures up to about 1000 K and pressures of maximum 70 atm the Chapman–Enskog equation provides good estimates of  $D$  [26].

From that equation it follows that:

$$D_{P_1, T_1} = \frac{P_0}{P_1} \left( \frac{T_1}{T_0} \right)^{3/2} D_{P_0, T_0} \quad (5)$$

The experimentally measured value of the diffusion coefficient of cyclohexane in oxygen at a temperature of 15 °C and at a pressure of 1 bar, is  $D = 0.0744 \text{ cm}^2/\text{s}$  [26]. With the help of Eq. (5) it is found that  $D = 0.0783 \text{ cm}^2/\text{s}$  at a temperature of 25 °C and at a pressure of 1 bar. For a bubble of 1.6 mm in radius, the characteristic time of the diffusion process is calculated from Eq. (4) to be  $\tau = 33 \text{ ms}$ . As is shown in Table 1, at this time point the saturation level of the bubble is about 77%. After  $3\tau$ , i.e. 100 ms, the saturation level of the bubble is about 97%.

This time is considerably less than the time the bubbles need to rise up to the center of the windows which is at least 500 ms. As a result, the cyclohexane pressure inside the gas phase of the bubble must be the vapor pressure at ambient temperature. The vapor pressure at 25 °C is 0.13 bar [20]. Therefore, the mole fractions of cyclohexane and oxygen in the bubble before the shock wave impact are expected to be 0.13 and 0.87, respectively. At a total initial pressure of 1 bar, this explosive mixture is nearly stoichiometric. Therefore, the oxygen bubbles in these experiments contain an explosive mixture of cyclohexane–oxygen already before the shock wave impact.

#### 4.3. Bubble ignition with short delay time

From experiments it is known [2–7,19] that the ignition of an explosive gas bubble takes place during the compression phase after the impact of the incident shock



wave. This is because the bubble reaches its highest temperature during the first compression. It is assumed that the appropriate temperature levels are reached for a time period long enough for the self-ignition of the bubble's gas phase.

A rough estimation of the time needed for this first compression after the shock wave impact, can be calculated by the Rayleigh equation. Under the assumption of total collapse of a spherical bubble that contains no gas phase (vacuum), the solution of the equation gives:

$$t_{\text{collapse}} \cong 0.9R_0 \sqrt{\frac{\rho_{\text{liquid}}}{P}} \quad (6)$$

where  $R_0$  is the initial diameter of the bubble before the impact,  $P$  the pressure of the shock wave during the compression, assumed to be constant over the compression phase and  $\rho_{\text{liquid}}$  the density of the liquid [27]. For a 3 mm bubble, and 100 bar magnitude of shock wave, the collapse time of such a bubble is about  $\tau_{\text{collapse}} = 12 \mu\text{s}$ . Taking in account that the bubbles are in reality not spherical and they contain a gas phase, this value correlates reasonably well with the observed short ignition delay ( $\tau_{\text{ign}} = 18\text{--}24 \mu\text{s}$ ). This corresponds to the explanations provided for similar ignition delays experimentally observed in other systems too. It is interesting to note at this point that bubbles of ( $\text{C}_2\text{H}_2 + 2.5\text{O}_2$ ) in water with a diameter of ( $2.7 \pm 0.3$ ) mm were ignited by shock wave loading as reported in [28]. The measured ignition delay,  $\tau_{\text{ign}}$ , in those experiments was about the same as in those of our experiments which gave the bubble explosion with a short ignition delay. This indicates the similarity of this ignition type in systems I and II.

Soot production is a distinctive characteristic in fuel-rich hydrocarbon systems. Such fuel-rich gas mixture can be produced by cumulative jet penetration during the interaction of the bubble with the incident shock wave. A theoretical analysis of the influence of soot production during the bubble explosion with short ignition delay was initiated in [29,30].

#### 4.4. Bubble ignition with long delay time

It was observed that a bubble breakage as a result of the impact from the shock wave was coupled with all the single bubble explosions that took place with the longer delay before ignition. A possible qualitative explanation for this phenomenon is given as follows.

It is known that the shock wave impact first results in a mechanical compression of the bubble. At some stage of this compression a penetrating jet may form. Photographic confirmation of this jet can be found in many publications, for example, in [17,23,31–35]. This penetration provides the inner part of the bubble with amounts of the surrounding liquid. The high temperature that the bubble has during its compression, allows some or all of this liquid to be evaporated into the gas phase of the bubble. Because of this evaporation, the temperature of the bubble's gas phase is reduced. Investigations revealed that the jet structure increases with solitary pressure wave amplitude and that the

jet entrains the whole gas bubble when the pressure wave intensity is above a certain limit [36].

If the jet formation is strong enough, its penetration results into the direct breakage of the bubble. In this case, the amount of liquid injected into the bubble's gas phase is significantly higher. Therefore, in the bubble, the temperature is essentially lower and the amount of cyclohexane is significantly higher, compared to the case without strong jet formation and bubble breakage. This consideration could provide the explanation why the new bubbles formed after the initial bubble shock-induced breakage, did not explode during their first oscillations. Either the bubble's gas mixture is not explosive because it reaches the fuel-overenriched area, or although it remains in the explosive area its temperature is not adequate to reach the self-ignition point.

In the experiments, a relatively long time was needed (more than 600  $\mu\text{s}$ ) before the bubble explosion finally took place. During this time, the pressure in the liquid is tending to reach lower levels of pressure (see Fig. 4), which means that the average bubble radius is becoming longer and therefore the gas temperature of the bubble is decreasing. Heat transfer phenomena on the bubble's surface (evaporation of cyclohexane, heat conduction) make this decrease more intense. Mass transfer phenomena in both directions, i.e. evaporation and condensation, are expected to play an important role for this processes too. It can be assumed that at the ignition time the bubble contains a very high fraction of gaseous fuel. This means that higher quantities of soot should be expected to be produced in this case, as compared to those from an explosion in the first oscillations.

It is interesting to note here that the ignition behavior of fuel-enriched hydrocarbon gas mixtures under relatively low temperature is still not a completely investigated area. Ignition under such conditions can have a much more complex mechanism with significantly different parameters as compared with the ignition in high temperatures [12]. In our situation, the processes are essentially further complicated by the heat and mass exchange phenomena and by soot formation that take place in the bubble. Further experimental and theoretical investigations in this direction are needed in order to reveal the mechanism of this new type of bubble ignition.

## 5. Conclusions

Two types of single bubble ignition in the system liquid cyclohexane–oxygen bubbles are observed. The first type is characterized by a bubble explosion in the first oscillation after the shock wave impact. A light emission with a duration of a few microseconds was observed during this explosion. This type of ignition corresponds to the bubble ignition reported for other systems too. The ignition delay of the second type were much longer, i.e. more than 600  $\mu\text{s}$ . This type of bubble ignition is reported for the first time. We suppose that strong heat and mass exchange phenomena

as well as the soot formation during the explosion strongly influence this type of ignition.

We observed both types of bubble explosion under the same initial conditions. This could be an indication that we worked in an intermediate area between two regimes where only one of the two bubble ignition types is possible. It is known that bubble explosions with short ignition delay can form self-sustaining shock waves, even in relatively short bubble columns. Although it seems impossible for bubble explosions with a long ignition delay to create a self-sustaining shock wave in columns of such lengths, it is at the present time not clear if such self-sustaining waves could indeed exist inside bubble columns of industrial scales. Since liquid hydrocarbon–oxygen bubble systems are very common in the industry, a clear need for further investigation of these phenomena is evident.

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