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Safety aspects of a bubbly medium inside a chemical reactor

K. Mitropetros*, P.A. Fomin, H. Hieronymus

Federal Institute for Materials Research and Testing (BAM), Unter den Eichen 87, D-12205 Berlin, Germany

Abstract

The possibility of explosion of a compressed oxygen bubble in liquid cyclohexane due to interaction with a spherical shock wave emitted by a nearby exploded bubble has been experimentally and theoretically investigated. Calculations for the explosion limits of a single bubble have been performed too. It is shown that in order to prevent bubble explosions inside a chemical reactor, the operating conditions (temperature and pressure) should be within a certain range.

Keywords: Cyclohexane; Oxygen; Shock wave

1. Introduction

Many processes in the chemical industry involve an organic liquid containing oxidizer bubbles. For example, the mixture: cyclohexane (liquid)–oxygen (bubbles) is used for the production of nylon. Wave phenomena in these systems, connected with shock induced bubble explosions have close connection with the performance and safety of chemical reactors. The first direct experimental registration of shock induced chemically active single bubble ignition in the liquid was performed in [1]. Shock wave initiation of chemically active liquid-bubble mixtures with a line of bubbles and with bubble columns were investigated in [2,3]. It was shown that a shock wave with amplitude above a critical value initiates the ignition of the medium behind the shock front.

There are two different mechanisms of single bubble ignition in chemically active systems. One of them is the increase of the gas pressure and the temperature owing to the bubble compression, caused by a shock wave in the liquid. This mechanism of bubble explosion was intensively discussed in the literature [1–5].

The second mechanism of bubble ignition is based on the penetration of a shock wave from the surrounding liquid into the bubble. The increase of the temperature and the pressure behind the shock wave inside the bubble can lead to bubble ignition. At normal initial conditions this mechanism of bubble ignition takes place only in relatively strong shock waves [6–8] with amplitudes, which are not reached in self-sustaining waves of compression in chemically active bubbly liquids, i.e. in bubble detonation waves [9–12]. If the gas inside the bubble has a relatively low pressure and temperature, the acoustical impedance of the gas is negligibly small compared to the corresponding value of the liquid. The acoustical impedance is given by ρc, where ρ is the density of the medium and c the velocity of sound [13,14]. Thus, there is an essential attenuation of the shock wave on the inter-phase boundary. As a result, the shock wave, which penetrates into the gas has low amplitude, which can be not enough to ignite the gas phase of the bubble.

A different situation is given when a shock wave impacts a bubble, which is already compressed. In this case the pressure, temperature, density and sound velocity inside the bubble are essentially higher, compared to the situation when the bubble is not compressed. Because of this, the difference in acoustical impedances between the gas of the bubble and the surrounding liquid is not high. As a result, the attenuation of the shock wave owing to penetration into the gas will not be high and the gas of the bubble could be ignited. The possibility of an explosion of an initially compressed bubble due to penetration of a shock wave into the bubble and the role of this kind of explosion in propagation mechanism of bubble detonation waves is still not adequately investigated.

In this work the shock induced behavior of a cluster of oxygen containing bubbles in liquid cyclohexane has been experimentally investigated. Explosions induced by bubble

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* Corresponding author. Tel.: +49 30 81 04 3426; fax: +49 30 81 04 1227. *E-mail address:* konstantinos.mitropetros@bam.de (K. Mitropetros).

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compression as well as explosions induced by shock waves penetrating into compressed bubbles were observed. Theoretical calculations were performed that explain the experimental observations.

Moreover, one of the important problems in the safety engineering of chemical reactors with bubble systems is to define those operating ranges of pressure and temperature that would not allow bubble explosions in the system. For this reason, the explosion limits of a bubble at different initial temperatures, pressures and for several amplitudes of incident shock waves have been calculated.

2. Experimental

A schematic of the autoclave, which was used in the experiments, is shown in Fig. 1. The autoclave has the form of a vertical cylindrical tube of 100 mm inner diameter and 1070 mm length. The bubble generator is installed at its bottom. 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 following positions are denoted in Fig. 1: (1) gas inlet, (2) gas outlet, (3) exploding wire, (4–7) pressure sensor positions, (8) liquid outlet and (9) gas inlet for the bubbles.

A shock wave in the liquid is generated by a gas detonation of the explosive acetylene–oxygen gas mixture above it. The liquid phase consisted of pure cyclohexane. The initial pressure of the mixture was 1 bar. All experiments were performed at room temperature. Detailed description of the experimental set-up can be found in [15].

3. Results and discussion

The shock wave that was generated in the liquid by the impact of the detonation in the gas phase was used to compress and consequently ignite the bubbles. In Fig. 2, oxygen bubbles in cyclohexane under shock wave impact are presented. Time zero corresponds to the moment when the shock wave enters the observation window. The incident shock wave can be observed on the frame at $t = 8\,\mu s$. After the shock wave passage, bubble compression follows. During this phase also jet penetration through the bubble can occur (e.g. see bubble no. 3). As a result of the bubble compression, ignition in some of the bubbles was observed (e.g. see Fig. 2, 33 μs).

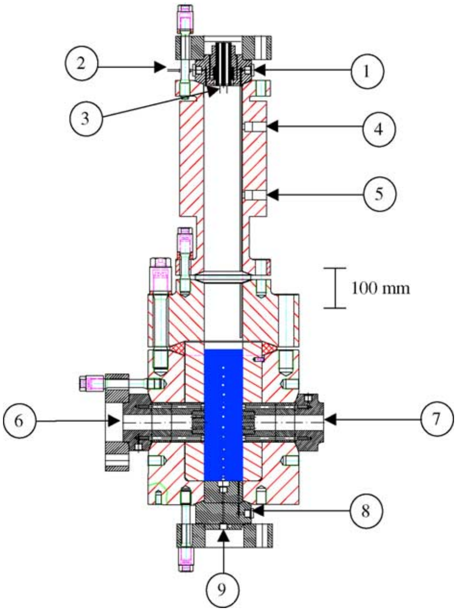

Fig. 1. Schematic of the autoclave.

It was observed that shock waves emitted by exploding bubbles were able to trigger bubble ignition of nearby compressed bubbles. For example, the bubble no. 2 ignition was initiated by the overlapping of two shock waves from nearby bubble explosions (Fig. 2, 35 μs). Then the shock wave of this explosion ignited the bubble no. 4 (Fig. 2, 40 μs). The fact that the gas phase of the bubble is compressed means

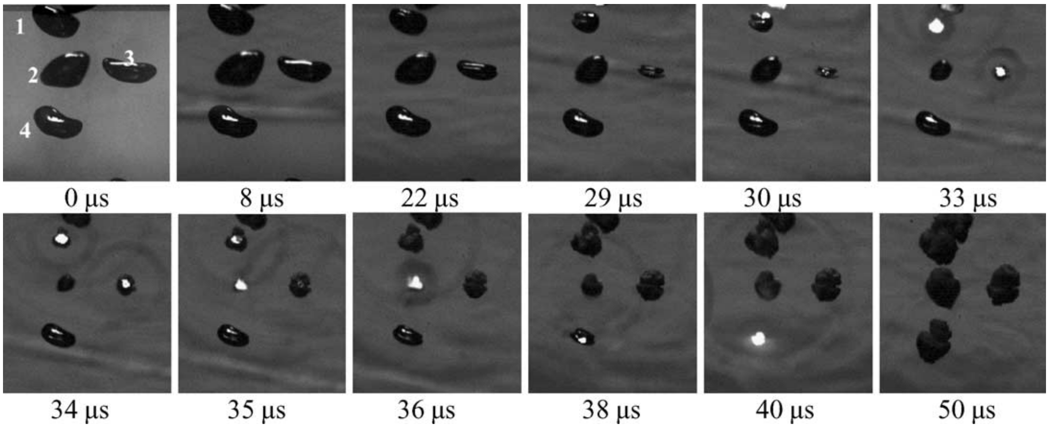

Fig. 2. Bubble ignition from a planar shock wave or from nearby bubble explosion. Initial equivalent diameter of the bubbles no. 1–4: 3.7, 4.1, 3.6 and 3.6 mm, respectively.

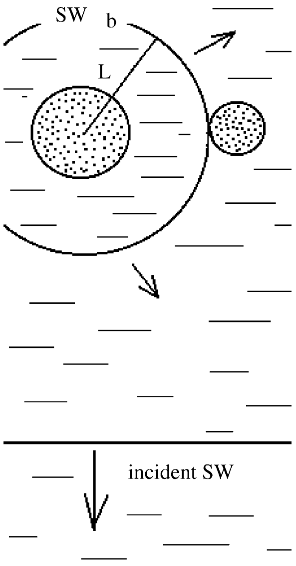

Fig. 3. Planar and spherical shock waves inside the liquid.

that the acoustical impedance is higher than the initial one in the bubble. Based on the experimental measurements of the bubble radius before the second shock wave impact, it is calculated that the increase of the acoustical impedance of the gas is about two orders of magnitude. The interaction of the shock wave with the preliminarily compressed bubble results into a new shock wave in the gas that ignites the bubble.

For the calculation, the following scenario was assumed. A planar shock wave with constant amplitude of 40 bar impacts two bubbles. As a result both bubbles start to shrink. During this process one of the bubbles explodes. At the moment of its explosion this bubble has a radius R_{ign}. The bubble explosion has a peak pressure P_{in}. The exploding bubble emits a spherical shock wave (SW_{b}) in the liquid with an amplitude initially equal to P_{ign}. This shock wave impacts on the surface of the second bubble (see Fig. 3). After the impact of the SW_{b} on the surface of the second bubble, a rarefaction wave in the liquid and a refracted shock wave inside the bubble ($\mathrm{SW}_{\mathrm{ref}}$) are created. This refracted wave has an initial peak pressure P_{ref} and initial propagation velocity u_{ref}.

At the moment of the shock wave impact, the bubble is nearly in mechanical equilibrium, i.e. the pressure inside this bubble is close to the pressure of the planar shock wave, 40 bar.

To calculate the explosion pressure P_{ign} inside the first bubble, the following theoretical model, presented in [4,5], is used. The bubble is assumed to be spherical. The bubble dynamics is described by the generally accepted Rayleigh-type equation [16]:

$$\beta\frac{\mathrm{d}^2\beta}{\mathrm{d}t^2}+\frac{3}{2}\frac{1}{\beta}\left(\frac{\mathrm{d}\beta}{\mathrm{d}t}\right)^2=\left(\bar{P}-\frac{P_{\mathrm{L}}}{P_0}\right)\frac{P_0}{\rho_{\mathrm{L}}R_0^2},$$

$$\bar{P}=\frac{1}{P_0}\left(P-\frac{2\sigma}{R}\right)-\frac{4}{\beta}\frac{\rho_{\mathrm{L}}\nu_{\mathrm{L}}}{P_0}\frac{\mathrm{d}\beta}{\mathrm{d}t}-3\frac{\rho_0}{\beta^3P_0}\frac{R_0}{C_{\mathrm{L}}}\left(1+\frac{P-P_{\mathrm{L}}}{B-P_{\mathrm{L}}}\right)^{-1/n}\frac{\mathrm{d}P}{\mathrm{d}\rho}\frac{\mathrm{d}\beta}{\mathrm{d}t}. \qquad (1)$$

Here R is the bubble radius, $\beta = R/R_0$ the dimensionless radius of the bubble, ρ and ρ_{L} are the gas and liquid density, P the pressure of the gas, ν_{L}, C_{L} and P_{L} are the kinematical viscosity, velocity of sound and pressure of the liquid, B and n are the constants of Tait equation of state of the liquid, t the time. Index “0” corresponds to the initial stage, the time moment $t = 0$ corresponds to the start of bubble compression.

The gas is assumed to be ideal:

$$\frac{P}{\rho}=\frac{\Re T}{\mu}. \qquad (2)$$

Here T and μ are the temperature and molar mass of the gas, $\Re$ the universal gas constant.

The point in time of the gas ignition t_{i} is determined by the conventional condition:

$$\int_0^{t_i}\frac{\mathrm{d}t}{\tau}=1, \qquad (3)$$

where τ is the induction period of a homogeneous gas mixture with constant parameters.

All chemical reaction proceeds in the gas phase. During the induction period the gas–vapour mixture is non-reactive, and thereafter it transits instantaneously into the state of chemical equilibrium. The chemical equilibrium is shifted as the bubble radius changes and liquid transforms into gas.

The molar mass of gas in the state of chemical equilibrium and specific internal energy of gas U are described according to the model of chemical kinetic [17–19]:

$$\frac{\rho}{\mu}\left(1-\frac{\mu}{\mu_{\max}}\right)^2\exp\left(\frac{E}{\Re T}\right)\left(\frac{\mu}{\mu_{\min}}-1\right)=\frac{AT^{3/4}}{4K_+}\left(1-\exp\left(\frac{-\Theta}{T}\right)\right)^{3/2}. \qquad (4)$$

$$U(T,\mu)=\left[\frac{3}{4}\left(\frac{\mu}{\mu_{\mathrm{a}}}+1\right)+\frac{3}{2}\left(\frac{\mu}{\mu_{\mathrm{a}}}-1\right)\frac{\Theta/T}{\exp(\Theta/T)-1}\right]\times\frac{\Re T}{\mu}+E\left(\frac{1}{\mu}-\frac{1}{\mu_{\min}}\right), \qquad (5)$$

where μ_{a}, $\mu_{\min}$, $\mu_{\max}$ are the molar masses of gas in the atomic, completely dissociated and completely recombined states, Θ the effective temperature of excitation of the vibrational degrees of freedom of the molecules, E the average energy of dissociation of the reaction products, A, K_+ are the rate constants of dissociation and recombination of the generalised reaction products. Parameters μ_{a}, $\mu_{\min}$, $\mu_{\max}$ are the functions of initial chemical compositions of the gas and can be calculated according to [17,18].

The constants of the model of chemical kinetic are: $E = 459$ kJ/mol, $A = 5 \times 10^{10}\ \mathrm{m^3/(kmol\,s\,K^{3/4})}$, $K_+ = 6 \times 10^8\ \mathrm{m^6/(kmol^2\,s)}$, $\Theta = 3000$ K. The algorithm for calculation of these constants was described in [17,18]. The constants $C_{\mathrm{L}} = 1284$ m/s, $\rho_{\mathrm{L}} = 7.8 \times 10^3\ \mathrm{kg/m^3}$ can be found in

[20]. The follow exponent of the Tait equation of state of liquid cyclohexane is used: $n = 7$. Thus, $B = \rho_L C_L^2 / n = 1837$ atm.

The bubble compression is assumed to be adiabatic. Adiabatic curve and adiabatic index γ of chemically equilibrium gas are described according to [21]:

$$\frac{\mathrm{d}T}{\mathrm{d}\rho} = -\frac{U_\mu \mu_\rho - (\Re T/\rho\mu)}{U_\mathrm{T} + U_\mu \mu_\mathrm{T}},$$

$$\gamma = \frac{\mathrm{d}ln\ P}{\mathrm{d}\ln\ \rho} = 1 - \frac{\rho}{\mu}\mu_\rho + \frac{\rho}{T}\left(1 - \frac{T}{\mu}\mu_\mathrm{T}\right)\frac{\mathrm{d}T}{\mathrm{d}\rho}. \qquad (6)$$

Derivatives U_μ, U_T, μ_ρ, μ_T are the explicit algebraic functions of T, ρ and can be obtained from (4) and (5). Eq. (6) can be used for chemically non-reactive gas too. In this case derivatives U_μ, μ_ρ, μ_T are equal to zero.

Eqs. (4)–(6) allow us to take into account a wide range of variation of the following parameters: (i) molar mass, (ii) adiabatic index and (iii) heat release of chemical reaction due to recombination and dissociation processes.

The jump of gas parameters at the time moment t_i is calculated from the energy conservation law, equation of state and chemical equilibrium. Bubble radius, gas density and parameters μ_a, μ_min, μ_max at this jump are constants.

Note, that compared with the classical Rayleigh equation for bubble dynamics, Eq. (1) takes into account the compressibility of the liquid and the energy losses owing to the viscosity and the acoustic radiation from the bubble. The difference between this model and other known models of wave propagation in a bubbly system is as follows. The adiabatic index of the gas is not assumed to be constant and the possibility of chemical reaction inside the bubble is taken into account.

The pressure P_ign inside the first bubble was calculated by the model described above to be in the order of 5×10^3 bar. The amplitude of the emitted spherical shock wave P_SSW by the bubble explosion, as function of the distance L between the wave front and the center of the bubble, can be described by the known equation for spherical acoustical waves [13]: $P_\mathrm{SSW} = P_\mathrm{ign}(R_\mathrm{ign}/L)$. Fig. 4 shows P_SSW according this equation as a function of the dimensionless distance L/R_ign.

The temperature inside the compressed, second bubble at the moment when the spherical shock wave impacted it was calculated too. For this calculation it was assumed that the bubble compression was adiabatic. Then by using the pressure of the incident shock wave as input, the temperature inside the compressed bubble was calculated by the model presented in [4,5] and described above. The temperature in the compressed bubble is then 585 K.

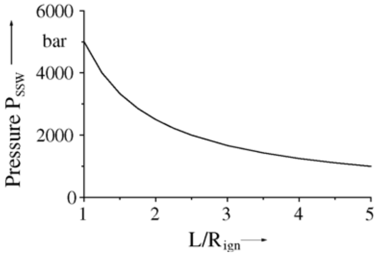

Fig. 4. Calculation for the pressure behind the secondary shock wave.

A one-dimensional simulation of the interaction of the $\mathrm{SW_b}$ with the surface of the second bubble was performed. In this simulation the parameters of the refracted shock wave were calculated only at the beginning of its propagation. For the simulation, the well-known theoretical description of interaction of a one-dimensional shock wave with inter-phase boundary was used, see e.g. [13,14]. For this description the equations of conservation of mass, momentum, energy and the equation of state are used. Apart from that, the pressure as well as the velocity of the gas and of the liquid on the inter-phase boundary are equal to each other.

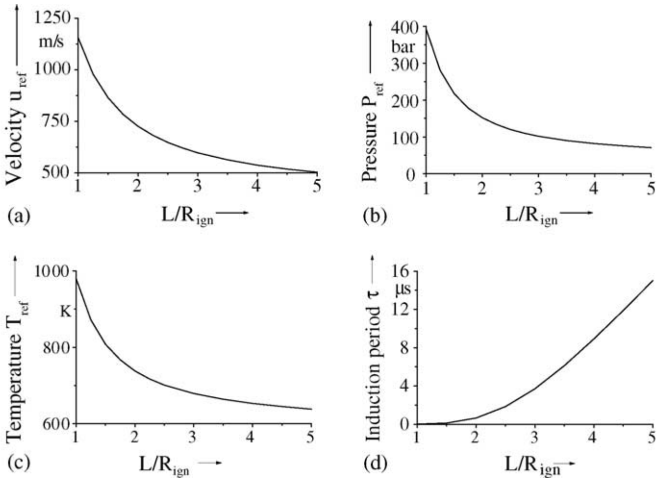

Fig. 5. Calculation for the velocity of the shock wave which penetrated the bubble (a), the pressure (b) and the temperature (c) behind such a wave and the induction period of the gas explosion (d).

Fig. 5 presents several calculated properties of the refracted wave. In Fig. 5a the calculated velocity of this wave u_ref is presented. The calculated initial amplitude of the wave P_ref and temperature T_ref behind the wave are shown in Fig. 5b and c, respectively. The calculated induction period of the gas explosion behind this wave τ, is presented in Fig. 5d. For this calculation the following equation was used: $\tau = AP_\mathrm{ref}^{0.5}\exp(\Sigma/\Re T_\mathrm{ref})$, where $\Sigma = 109$ kJ/(kmol), $A = 5.1 \times 10^{-11}$ s $\mathrm{Pa}^{0.5}$ [4,5].

A direct conclusion from Fig. 5 is that, provided the two interacting bubbles are close enough, the shock wave that penetrates the second bubble can ignite it. More precisely the calculation shown in Fig. 5d means that the pressure and the temperature behind the refracted wave can be so high that the induction period for the ignition is less than the time necessary for the bubble to change its radius (i.e. a few µs). This is the explanation of the "instantaneous" ignition that was observed in the experiments (see Fig. 2 at 35 µs). For illustration, if L/r_ign, is less than 2, then the induction period

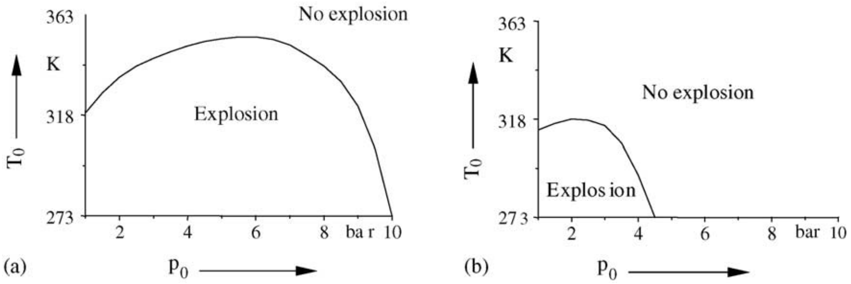

Fig. 6. Calculated explosion limits of a single bubble in the plane of initial pressure and temperature at different amplitudes of shock waves (40 bar (a) and 20 bar (b)).

is less than 0.65 μs, i.e. the second bubble would seem to ignite instantaneously at a μs scale.

In the following paragraph we consider the case of shock-induced compression of a single bubble. In this situation the bubble is under relatively low initial pressure and temperature. Thus there is bubble compression due to the shock wave but the penetration of the shock wave into the bubble is negligible. For this case in Fig. 6 the calculated explosion limits of a single bubble are presented. For this calculation it was assumed that if the duration of the first bubble compression is less than the induction period of the gas explosion, the bubble does not explode. The duration of the bubble compression as well as the induction period of the gas explosion inside the same bubble were calculated by the model described in [4,5] and described above. The calculation is presented as a function of the initial temperature and initial pressure of the system and for two different amplitudes of incident shock waves. In Fig. 6 it can be seen that for a certain amplitude of incident shock wave, a line in the plane of initial pressure and temperature exists, above which no bubble explosion can take place.

The influence of inter phase mass and heat transfer processes through the bubble's surface on the explosion limits of a single bubble, has been analyzed. As a first approximation, the bubble surface is assumed to be spherical, without disturbances. The following formula is used to estimate an upper limit for the mass of the liquid that is evaporated at the bubble's surface [22]: $L\,\mathrm{d}M/\mathrm{d}t = q_{\mathrm{w}}S$. Here M is the mass of gas inside the bubble, q_{w} the specific (per unit of surface square) heat flux from the gas to the bubble surface, S the square of the bubble surface ($S = 4\pi R^2$), L the heat of evaporation of the fuel. The heat flux is calculated by the formula [23]: $q_{\mathrm{w}} = \sigma(T - T_0)$. The heat-exchange coefficient σ can be estimated according to [23,24]: $\sigma = \lambda\pi^2/3R$, where λ is the thermal conductivity of gas. Parameters L and λ can be found in [20].

The calculations show, that the influence of inter phase transfer processes on the explosion limits of a single bubble is negligibly small. The calculation shows that during the first bubble oscillation, the mass of the fuel that is evaporated into the bubble is only a few percent of the initial mass of the bubble's gas. This mass addition practically does not influence the temperature and pressure of the gas, the induction period of chemical reaction and the duration of the first bubble oscillation.

The results presented in Fig. 6 support the following conclusion. In order to prevent bubble explosions in a chemical reactor, the operating temperature should be relatively high. The explanation of this follows. It is known, that the adiabatic compression of a gas strongly depends on the adiabatic index. The adiabatic index of hydrocarbon–oxygen gaseous mixtures is reduced if the concentration of hydrocarbons is increased. If the temperature of the bubbly mixture increases at a constant pressure, the cyclohexane concentration in the bubble increases too and the adiabatic index of the gaseous mixture decreases. Thus, at the same stage of compression the gas temperature will be higher inside that bubble, which was initially under lower initial temperature.

4. Conclusions

In the present work safety related aspects of shock induced phenomena inside a cluster of oxygen containing bubbles in liquid cyclohexane has been experimentally and theoretically investigated. Explosion due to bubble compression was experimentally observed. Direct bubble ignition due to refraction of a shock wave impact on compressed bubbles was observed too.

The calculated parameters behind the refracted shock wave inside a chemically active bubble have been presented. According to the calculations, the ignition delay behind the refracted shock wave is shorter as the time, which is necessary for the bubble to change its radius considerably due to shock wave impact. This means, that a refracted shock wave inside a compressed bubble can ignite it practically instantaneously.

The observed phenomenon that bubble explosions can ignite nearby bubbles indicates that an automatic process of synchronization of bubble explosions is possible. This result provides a first step to a better understanding of the mechanism of self-sustaining waves inside bubbly media.

Additionally, the calculated explosion limits of a single chemically active bubble under shock wave impact are presented. The calculations show the possible range of conditions for a safe operation of a chemical reactor containing a bubbly liquid.

Acknowledgements

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