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Mechanism of the sonochemical production of hydrogen

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ABSTRACT

It has been long recognized that propagation of an ultrasonic wave in water results in hydrogen production. The chemical effects of ultrasound (sonochemistry) originate from acoustic cavitation, that is, the formation, growth and implosive collapse of microscopic bubbles in liquid irradiated by ultrasound wave. Enormous temperatures and pressures are generated within the bubbles at the collapse, making each bubble as a microreactor within which typical flame reactions occur. The combustion in the cavitation bubbles yield species such as $^{\bullet}OH$, $H^{\bullet}$, O, $HO_2^{\bullet}$ and others. Although H_2 is the most molecular product of water sonolysis, the mechanism of its production is until now not understood and the most reported suggestions are controversial. In this paper, a comprehensive numerical work was carried out, for the first time, to explain the mechanism of ultrasound induced generation of H_2 in water. Computer simulations of chemical reactions occurring inside a bubble oscillating in water irradiated by an ultrasonic wave have been performed for different conditions. A kinetics mechanism of 25 reversible chemical reactions was proposed for studying the internal bubble-chemistry. The numerical simulations have evidenced the formation of H_2 as well as other products such as O_2, H_2O_2, $^{\bullet}OH$, $H^{\bullet}$, $HO_2^{\bullet}$ and O in the bubble during implosion. In all cases, H_2 was the main product formed in the bubbles at appreciable amount. Basing on the simulation results and using material balance for hydrogen in the gas and liquid phases, the production rate of H_2 in each phase has been quantified. The conclusion was that the main source of H_2 production during water sonolysis is the gas phase of the bubbles through the reaction $H^{\bullet} + {}^{\bullet}OH \leftrightarrow H_2 + O$.

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Introduction

Approximately 80% of the present world energy demand comes from fossil fuels [1]. Unlike fossil fuels, hydrogen gas (H_2) burns cleanly without emitting any environmental pollutants [2]. In addition, H_2 is also abundantly available in the universe and processes the highest energy content per unit of weight (i.e. 120.7 kJ/g) compared to any of the known fuels [1]. H_2 is considered to be the energy carrier of the future [3] and could have an important role in reducing environmental emissions. Several techniques have been used in the past for producing hydrogen. Steam methane reforming is the most widely used method for producing it [1,4]. Alternative ways could be used to generate H_2 such as ethanol gasification [5], water electrolysis [4,6], biological photosynthesis [7,8] and photocatalysis [9–11]. More recently, sonolysis of water have been successfully used for producing hydrogen [12–14]. Moreover, ultrasonication assisted hydrogen production from catalysis [15], photocatalysis [16–18], digestion sludge [19–21] and anaerobic fermentation of wastewater [22] have proven their improvement potential compared to each isolated technique (without ultrasound).

Since 1962, it has been recognized that propagation of an ultrasonic wave in aqueous solution (simply known as sonolysis) containing a solute can results in an oxidation process, which is called sonochemistry [23]. Besides this process, it was well established that H_2O_2 and H_2 are the main products of pure water sonolysis and the ratio of H_2 to H_2O_2 production is ~1.25 [13,14,24]. The generation of H_2 by water sonolysis does not arise from the direct interaction between acoustic wave and water but it produces upon acoustic cavitation, that is, the formation, growth and violently collapse of microscopic bubbles (filled with water vapor and dissolved gases) in a liquid medium irradiated by an ultrasonic wave (Fig. 1) [25]. The rapid collapse of these microbubbles is nearly adiabatic, making each bubble as a powered microreactor inside which temperature of several thousands of Kelvin and pressure of several hundreds of atmospheres are reached [26]. Under these enormous conditions, high temperature combustion-chemistry occurs in the bubble [27]. In fact, the trapped molecules in the bubble (water vapor, gases and vaporized solutes) can be brought to an excited-state and dissociate. As results, reactive species such as •OH, HO_2•, H•, O and H_2O_2 are created from H_2O and O_2 dissociation and their associate reactions in the bubble [28]. These chemical products may diffuse out of the bubble and dissolve in the surrounding liquid [29]. One of the most products of water sonolysis is hydrogen (H_2). It may be formed at rate of 10–15 μM min^{-1} [13,14,24], which is much higher than that obtained by photocatalysis (~0.035 μM min^{-1} [16]).

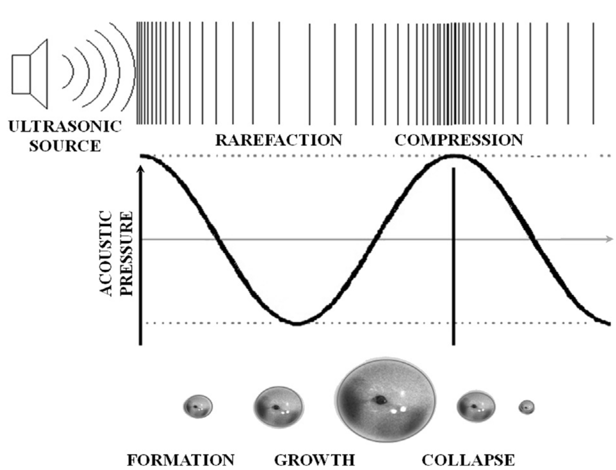

Fig. 1 – Dynamics of acoustic cavitation bubble.

The mechanism of H_2 production from water sonolysis is until now under discussion. A number of researches [24,30,31] postulated that the major part of H_2 is produced in the gas phase of the bubble and diffused to the solution whereas others [32,33] indicated that H_2 is formed only at the liquid shell of the bubble by the recombination of hydrogen radicals diffused from the bubble (H•+H•↔H_2). However, to the best of our knowledge, the mechanism of hydrogen production from water sonolysis is until now not understood. The present work is a contribution to the study of the mechanism of hydrogen generation during water sonolysis. A series of numerical simulations of chemical reactions occurring in the interior of an argon bubble oscillating in liquid water irradiated by an ultrasonic wave have been performed for various conditions. A kinetics mechanism of 25 reversible chemical reactions was postulated for studying the internal bubble-chemistry. On the basis of the simulation results, material balance for hydrogen in the gas and liquid phases has been carried out to evaluate the importance of each of the controversial reported suggestions and then determine the mechanism of H_2 generation during water sonolysis.

Model and computational methods

The theoretical model used in the present numerical simulations have been described in our previous works [34–36]. It combines the dynamic of single bubble in acoustic field with chemical kinetics consisting of series of chemical reactions occurring in the bubble at the collapse phase. The following is a brief description of the model.

Bubble dynamics model

A gas and vapor filled spherical bubble isolated in water oscillates under the action of a sinusoidal sound wave. The temperature and pressure in the bubble are assumed to be partially uniform and the gas content of the bubble behaves as an ideal gas [37]. The radial dynamics of the bubble is described by the Keller equation that includes first order terms in the Mach number M=$\dot{R}$/c [38,39]:

$$\left(1-\frac{\dot{R}}{c}\right)R\ddot{R}+\frac{3}{2}\left(1-\frac{\dot{R}}{3c}\right)\dot{R}^2=\frac{1}{\rho_L}\left(1+\frac{\dot{R}}{c}+\frac{R}{c}\frac{d}{dt}\right)\left[p-p_\infty-\frac{2\sigma}{R}-4\mu\frac{\dot{R}}{R}+P_A\sin(2\pi ft)\right] \quad (1)$$

in this equation dots denote time derivatives (*d/dt*), *R* is the radius of the bubble, *c* is the speed of sound in the liquid, ρ_L is

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