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Short Communication

Importance of acoustic shielding in sonochemistry

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Abstract

It is well known that sonochemistry is less efficient at high acoustic intensities. Many authors have attributed this effect to decoupling losses and shielding of the acoustic wave. In this study we investigate both phenomena for a 20 kHz ultrasound field with an intensity ranging from 40 to 150 W/cm^2. Visualization of the bubble cloud has demonstrated that the void fraction below the ultrasound horn increases more than proportional with increasing power input. Nevertheless, the energy coupling between the horn and the liquid remains constant; this implies that decoupling losses are not reinforced for larger bubble clouds. On the contrary, microphone measurements have shown that due to the larger bubble cloud a substantial part of the supplied energy is lost at high power inputs. In striving towards more efficient sonochemistry, reduction of shielding appears as one of the major challenges.

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Keywords: Sonochemistry; Ultrasound power; Bubble cloud; Sound attenuation; Decoupling

1. Introduction

Ultrasonic irradiation has proven to be a versatile and promising tool for the development of new processes in the chemical industry [1,2]. Its unique character predominantly arises from acoustic cavitation, i.e. the growth and subsequent adiabatic collapse of a microscopic cavity in a liquid, leading to a momentary increase of temperature and pressure. These extreme local conditions can cause bond breakage and free radical formation, thereby providing an alternative route for inducing chemical reactions (sonochemistry).

It seems evident to increase the sound field intensity to enhance sonochemical reaction rates. The intensity can be increased either by reducing the radiating surface area or by increasing the power supplied to the system. In case of an existing experimental set-up, the power input is generally used to adjust the acoustic intensity. It has been reported that the sonochemical decomposition of organic solutes and the oxidation of potassium iodide increase linearly with increasing acoustic intensity, using low-frequency ultrasound (20–60 kHz) of relatively low intensity. At high intensities, however, an increase in intensity leads to a relatively small increase or even a decrease in the sonochemical efficiency [3–8]. Wu et al. has even demonstrated that in a single experimental set-up the dependency on acoustic intensity can vary for different reactions [9].

Several explanations have been proposed in the literature to account for the remarkable effect of intensity. With increasing power input the sound field penetrates further into the liquid and consequently, the sonochemical active zone covers a larger region. This effect has been confirmed by previously reported sonogenerated chemiluminescence studies [10,11]. On the contrary, the increased number of cavitation bubbles can absorb and scatter the sound wave, thereby weakening the acoustic field substantially (shielding) [12,13]. Furthermore, these bubbles change the acous-

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tic impedance of the medium and as a consequence the energy transfer efficiency from the ultrasound emitter to the medium can decrease. This phenomenon is referred to as decoupling losses. The aim of this study is to investigate to what extent decoupling losses and shielding effects cause sonochemistry to be less efficient at elevated intensity. Previous work has demonstrated that for intense sonication at 20 kHz a small increase in hydrostatic pressure results in a higher sonochemical reaction rate [14]. To confirm that at these conditions this sonication system also suffers from less efficient sonochemistry at high acoustic intensities, the oxidation of potassium iodide has been studied at different power inputs. Calorimetric measurements have been performed to determine the acoustic energy supplied to the liquid and to investigate the occurrence of decoupling losses [15]. Laser light scattering has been used to visualize the bubble cloud and microphone measurements have been done to study the effect thereof on sound attenuation. It should be emphasized that in contrast to several other studies, the images of the bubble cloud are not used for detecting sonochemical activity, yet for investigating shielding effects [11,16].

2. Experimental

An ultrasonic generator with variable power output (Sonics and Materials Inc., VC750) was used for producing ultrasound with a frequency of 20 kHz. The piezoelectric transducer was coupled to the liquid with a 13 mm diameter full wave titanium horn. In all experiments, the horn was inserted at the top of the reactor, fixed at its nodal point and immersed 20 to 30 mm below the surface of the liquid. Previous work has demonstrated that for these sonication conditions a maximum in reactivity is observed at a hydrostatic pressure of 5 bar [14]. Therefore, all measurements were performed for water (Millipore filtered) pressurized at 5 bar using argon (grade 5.0, Hoek Loos B.V.). Each measurement was performed in consecutive order, starting with the lowest intensity.

2.1. Oxidation of potassium iodide

The effect of acoustic intensity on sonochemical reactivity was studied by means of the oxidation of potassium iodide in aqueous solution. For aqueous systems, cavitation results in the formation of OH-radicals. These radicals can react with I^- to give I_2, which reacts further in the presence of an excess of I^- to yield I_3^-. The concentration of I_3^- was determined spectrophotometrically at 352 nm [17].

The sonochemical experiments were performed in a 175 mL high-pressure vessel with a diameter of 50 mm. The temperature inside the reactor was controlled externally using a thermostatic bath and a Pt-100 temperature sensor (Fig. 1). To monitor the course of the reaction, the reactor was connected to a small high-pressure view cell with sapphire windows (SITEC-Sieber Engineering AG, 740.2095), which was placed within a UV–visible spectrophotometer (Thermo Electron Corporation, Genesys 5). The sonicated solution was circulated through the view cell using a HPLC pump (Jasco Inc., PU-2086 Plus).

The reactor was filled with 150 mL of 0.1 M potassium iodide solution (KI $\geqslant$ 99.0%, Sigma–Aldrich) and the temperature inside the reactor was controlled at 20 °C. To saturate the solution with argon and to remove all air present,

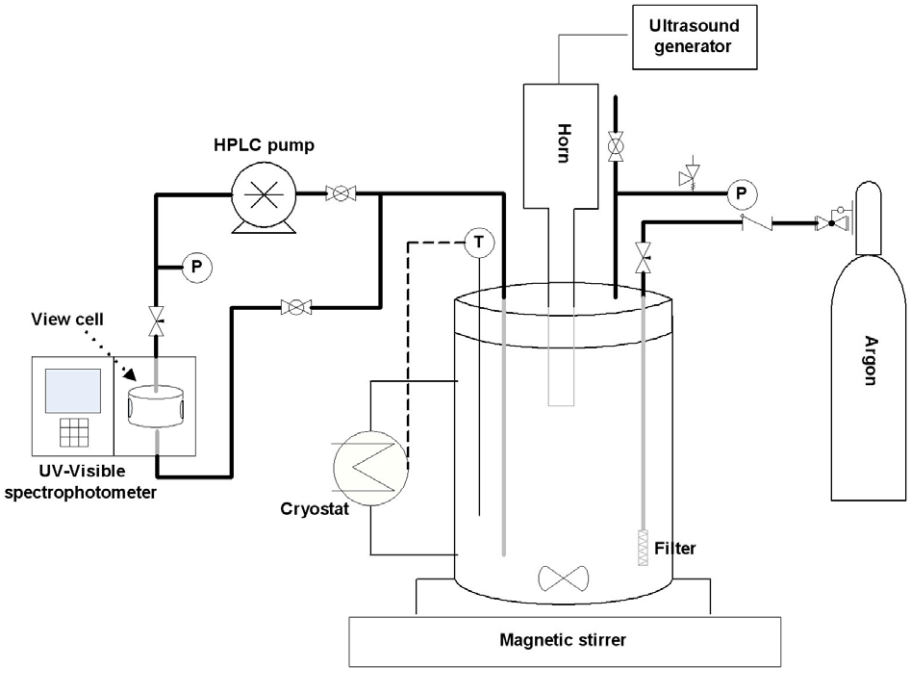

Fig. 1. Experimental set-up for sonochemical experiments.