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Characterization of the activity of ultrasound emitted in a perpendicular liquid flow using Particle Image Velocimetry (PIV) and electrochemical mass transfer measurements

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ABSTRACT

The present work is dedicated to the study of the interactions between a liquid circulation and a perpendicular acoustic wave propagation. A specific experimental setup was designed to study one transducer operating at 20 kHz, with the help of electrochemical mass transfer measurements combined with Particle Image Velocimetry (PIV) determination. Electrodes were located on the wall opposite to the acoustic emission. Experiments were performed for various Reynolds numbers: from 0 to 21700 (different liquid flow rates and viscosities). Both PIV and electrochemical measurements methods were found to be relevant, and had delivered complementary information. Even if PIV showed that the plume due to streaming was highly deflected by the additional flow, electrochemical measurements showed that there was still an activity, higher than in silent conditions, on the wall facing the transducer. Thus the ultrasound contribution remained noticeable on the surface opposite to the transducer even for a disturbed hydrodynamic environment due to the presence of a liquid circulation perpendicular to the wave propagation.

1. Introduction

Through mechanical, thermal and chemical effects induced by wave propagation, power ultrasound appears as a promising tool in an increasing number of industrial applications such as extraction [1], emulsification [2], cleaning [3]... The main effect is already known to be the generation of cavitation bubbles, responsible for mass transfer enhancement, radical formation and interface cleaning [4]. All those phenomena often contribute to an enhancement of the chemical reaction kinetics. Nevertheless, the dissemination of large scale functional unit in production is slowed down by difficulties in design: major problem consists in heterogeneities in the acoustic power distribution in the reaction volume. In the last decades, different methods have been developed for the determination of active zones. Some of them give a global quantification of the acoustic activity such as calorimetry [5], chemical radical dosimetry [6], whereas local information are given by determination of mass transfer [7] or laser visualizations (Tomography [8] and Particle Image Velocimetry [9]). In papers available in the literature, acoustic activity is usually described in an initially motionless fluid bulk, i.e. in the case of the liquid motion comes only from the transducer activity [7]. Studies are then focused in the vicinity of the transducer horn, where the fluid flow is induced by the wave propagation [10]. However, numerous large scale applications of ultrasound involve an additional liquid flow inside the acoustic field. For example, in the case of displacements of an acoustic cleaning tool at a surface for ship hulls cleaning applications [3], or in the case of circulation of reactants into a sonoelectroreactor [11]. In this last example, electrodes are located face to face with the transducers, and the fluid circulates perpendicularly to the wave propagation. In those situations, two methods are relevant to provide useful information on local acoustic activity. The first one is the electrochemical measurement of mass transfer phenomena by cyclic voltammetry using the well-known quasi-reversible redox couple $Fe(CN)_6^{3-}/Fe(CN)_6^{4-}$. The second one is Particle Image Velocimetry (PIV) for velocity fields determination. PIV is a non-intrusive optical method which enables to obtain 2D velocity vectors fields in a plane of a flow [12]. Two successive laser pulses illuminate a sheet of a particles seeded flow, so that a camera can record 2 consecutive images of particles in the plane. Then, cross-correlation between subdivisions of these 2 images enables to calculate the particles displacements field during the delay time between the 2 recordings. Knowing this delay time and the spatial

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calibration, a velocity vector field can be constructed. Both techniques enable to describe and quantify the fluid convective motion into sonoreactors, the so-called ultrasonic wind [13,14], whereas mass transfer measurements allow a local evaluation of all velocity gradient's contribution at the electrode surface. The main contribution of stirring at the electrode surface facing the ultrasonic horn accounts for as much as 90% from asymmetric cavitation [15]. The present study takes into account the influence of a liquid flow perpendicular to the wave propagation in a dedicated experimental set-up allowing various flow rates and viscosities (Reynolds numbers varying from 0 to 21,700) and equipped with transducer operating at 20 kHz. Both PIV and electrochemical measurements were carried out to quantify the effect on the ultrasound activity of the liquid flow rate increase. It is important to note that, in this paper, the possible influence of natural convection is included in the term "streaming".

2. Materiel and methods

A dedicated set-up was especially designed to investigate effect on ultrasound activity of a liquid flow perpendicular to the wave propagation. A rectangular cross-section polypropylene cell (35 mm × 40 mm and 400 mm length) of measure was included (Fig. 1). Transparent PMMA front and top windows permitted the laser visualization. The liquid was contained in a thermostated tank (25 L) and controlled flow circulation was supplied by a centrifugal pump. The ultrasound probe was embedded in a flange fixed at the bottom face of the cell. Concerning the electrode used for mass transfer determination, it was embedded in a flange fixed at the top face of the cell. Three different locations of the electrode were possible, corresponding to the so-called local observation areas on the Fig. 1.

For all experiments carried out at 20 kHz, a probe (Sonics & Materials, Danbury USA) was used as the ultrasound source (amplitude of 60%, transmitted power of 51 W).The titanium horn we used (to transmit ultrasound to the liquid media) had a radiating face which was a truncated disk (25 mm in length and 22 mm in width, cf. Fig. 1). The probe was located such as the longest side was in the main liquid flow direction. The working electrode was a platinum disk (1 mm in diameter, 7.85×10^{-3} cm^2) embedded in a glass tube and polished down to 1/4 μm granulometry. Its surface was located opposite and parallel to the horn face. A simple platinum wire was used as a quasi reference electrode, and the counter electrode consisted of a platinum plate designed and placed so as to be outside the ultrasonic field. The redox couple ($Fe(CN)_6^{3-}/Fe(CN)_6^{4-}$) was used for the mass transfer measurements. The solution was potassium ferri/ferrocyanide (5×10^{-3} mol L^{-1}) in sodium hydroxide (0.2 mol L^{-1}). By adding specific quantities of Polyethylene glycol, the solution's kinematic viscosity varied from 8.2×10^{-7} to 15.6×10^{-7} m^2 s^{-1}. Mass transfer measurements under sonication were taken using the electrodiffusion method. For this purpose, sonoelectrochemical voltammograms of the ferri-ferrocyanide reversible couple on a stationary working electrode were plotted. A typical voltammetry curve exhibits a sigmoid current response yielding a signal plateau corresponding to mass transfer limited current. This value of current density under ultrasonic stirring includes a steady state component and a time-dependent component (oscillation around the average plateau current value). Agitation at the electrode surface was quantified by electrochemical measurements and quoted in terms of equivalent flow velocity, which was deduced from velocity gradients at the electrode surface [13]. In the specific case of a tangential flow to the electrode surface (due to recirculation on the surface facing the transducer, and due to the additional flow), a parietal velocity V_p was calculated by means of the following equation [16]:

$$V_p = \frac{1}{(0.6\,nFC_{sol})} D^{-4/3}\,\nu^{1/3} j_D^2\, d^{2/3}\, x^{1/3} \quad (1)$$

where D is the diffusion coefficient (m^2 s^{-1}), C_{sol} is the concentration of the electroactive species (mol m^{-3}), ν is the kinematic viscosity (m^2 s^{-1}), n is the number of electrons transferred, d is the electrode diameter (m), F is the Faraday number, j_D is the current density (A m^{-2}) and x, in this case, represents the distance between the electrode center and the beginning of limiting diffusion layer [16]. This parietal velocity was compared and found to be in agreement with the ones obtained from flow rate and Particle Image Velocimetry (PIV) measurements in silent conditions [17].

The experimental setup used for PIV measurements was composed of a dual beam pulsed Nd:YAG laser (532 nm) paired with a camera (PIVCAM 13-8, 12 bits). Optical components (cylindrical and spherical lenses, prism) allowed to spread the laser beam as a sheet which was redirected toward the liquid. A synchronizer trigged both the laser and the camera. At a given frequency, two laser beams were flashed and were separated with a very short time interval, called pulse delay Δt. This last one was set at 100 ms or 300 ms (depending on the operating conditions). For each flash, an image of the flow was recorded by the camera. The liquid media was seeded with tracer particles (hollow glass beads, 8–12 μm diameter). For each operating condition, a set of 100 pairs of images were recorded. The whole pre-processing and processing of the images were achieved using the PYV software developed at the Femto-st institute [18–20]. First, images were pre-processed. PYV software allowed us to correct the relative displacement between two successive pairs of images, and which was due to pump's vibrations. Indeed, recorded images did not have the same spatial origin. Using the classical PIV correlation process between a reference image and the rest of the images, a relative displacement was calculated. Images were then translated of the displacement's value and saved. With this software we also corrected image orien-

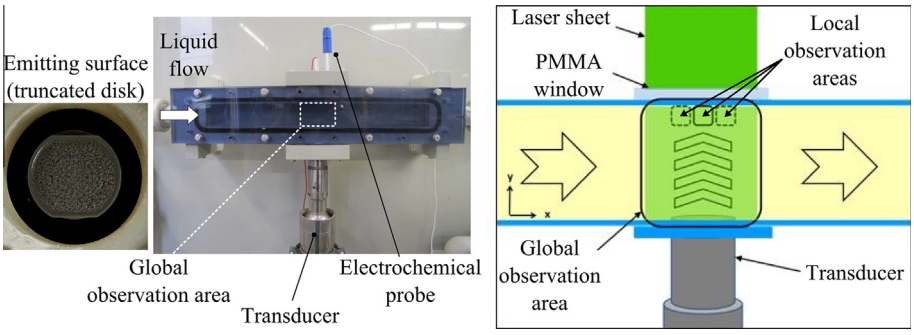

Fig. 1. Test cell for electrochemical and PIV measurements.