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Characterization of HIFU transducers designed for sonochemistry application: Acoustic streaming

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ABSTRACT

Cavitation distribution in a High Intensity Focused Ultrasound sonoreactors (HIFU) has been extensively described in the recent literature, including quantification by an optical method (Sonochemiluminescence SCL). The present paper provides complementary measurements through the study of acoustic streaming generated by the same kind of HIFU transducers. To this end, results of mass transfer measurements (electrodiffusional method) were compared to optical method ones (Particle Image Velocimetry). This last one was used in various configurations: with or without an electrode in the acoustic field in order to have the same perturbation of the wave propagation. Results show that the maximum velocity is not located at the focal but shifted near the transducer, and that this shift is greater for high powers. The two cavitation modes (stationary and moving bubbles) are greatly affect the hydrodynamic behavior of our sonoreactors: acoustic streaming and the fluid generated by bubble motion. The results obtained by electrochemical measurements show the same low hydrodynamic activity in the transducer vicinity, the same shift of the active focal toward the transducer, and the same absence of activity in the post-focal axial zone. The comparison with theoretical Eckart's velocities (acoustic streaming in non-cavitating media) confirms a very high activity at the "sonochemical focal", accounted for by wave distortion, which induced greater absorption coefficients. Moreover, the equivalent liquid velocities are one order of magnitude larger than the ones measured by PIV, confirming the enhancement of mass transfer by bubbles oscillation and collapse close to the surface, rather than from a pure streaming effect.

1. Introduction

In the first part of this study [1], we have described the cavitation distribution in a sonoreactor equipped with High Intensity Focused Ultrasound (HIFU) and quantified them with an optical method (SCL). The use of HIFU enables generation of very intense acoustic intensities at high frequencies, which is not possible with classical flat transducers. This large density of acoustic energy had already paved the way for very interesting medical applications, allowing specific innovations in therapeutics [2,3]. More recently, prospects have emerged in surface treatments such as selective ablation of polymers [4], study of cavitation and hydrodynamic effects on the selective stripping of acrylic coatings, selective polymerization known as "acoustic masking" [5,6], or modification of compactness and ions repartition of polypyrrole coatings electrosynthesized under ultrasonic irradiation and selective oxidation of pyrrole on oxydable metals.

Characterization of this kind of transducer is rather complex due to the presence of cavitation bubbles in the acoustic field. As predictive tools are not available for cavitation phenomenon, experimental studies had to be conducted to describe their specific behavior. In a previous paper [1], the dynamics of cavitation bubbles can be defined in two modes: "stationary bubbles" (bubbles trapped in the pressure antinodes of the standing wave) and "moving bubbles" (bubbles in motion in the acoustic field). The present work concerns the acoustic streaming generated by HIFU.

The flows generated by ultrasound are nearly always present in all sonoreactors, along with cavitation and other induced effects. Such flows are well known to enhance mass transfer in processes limited by diffusion [7,8], to favor heat exchanges [9,10] or indeed to perform mixing or micromixing [11,12]. Wave propagation in a viscous fluid induces a transfer of energy between the acoustic wave and the propagation media, leading to a large scale flow commonly called "acoustic streaming" [13]. In a non-cavitating media,

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two kinds of acoustic streaming can be seen: Eckart's streaming (or quartz wind), generated in the bulk by the Reynolds tensions resulting from wave absorption in a viscous media, and Rayleigh streaming where the Reynolds tensions act on the hydrodynamic limiting layer in the vicinity of the reactor wall [14]. In cavitating media (heterogeneous media), a variety of factors are involved in global agitation. The present work is based on experimental measurements using optical method (Particle Image Velocimetry) and electrochemical method to describe the global agitation generated by HIFU.

2. Experimental details

2.1. Instrumentation

All experiments were conducted using two composite HIFUs designed by IMASONIC (Besançon, France). The first operates at 3 MHz (Tfc3000) with a 40 mm geometrical focal length, while the second operates at a frequency of 750 kHz (Tfc750) with a 100 mm geometrical focal length. Transducers were set on the vessel's bottom. The geometrical focal corresponds to the center of spherical cap of the emitting surface characterized by its radius [15]. The reactor (750 mL) consists of a double walls cylindrical Pyrex vessel (92 mm diameter) equipped with a displacement system with four degrees of freedom [1]: three translations guided by micrometric screws and one rotation axis to avoid reflections toward the transducer.

2.2. Operating procedures

In the present work, streaming velocities were measured by two methods. Particle Image Velocimetry (PIV) is an optical visualization method dedicated to velocity measurements by determining particle displacement over time using a double-pulsed laser technique [16,17]. The liquid media was seeded by tracers, which have a density close to water (microparticles of Rilsan 30 μm). A light sheet, generated by a laser Continuum Nd-Yag (532 nm) and an optical system with cylindrical lenses, illuminates the acoustic axis plane. Particle locations are recorded in this plane using a CDD camera (Sensicam PCO 12 bits, 1280 × 1024 pixels). A fraction of a second later, a second image of the particle was taken and from these two images, analysis algorithms allowed to obtain the particle displacements for the entire flow region mapped. For each experiment, 50 pairs of images were recorded and post-treated using INSIGHT 3D software.

The second method was the determination of mass transfer by cyclic voltammetry using the well-known quasi reversible redox couple $Fe(CN)_6^{3-}/Fe(CN)_6^{4-}$. The work of Coury et al. [18–20] and Compton et al. [21] investigated mass-transfer phenomena under sonication. Sonication decreases the diffusion layer thickness δ (m) and increases the limited current of diffusion $|\vec{j}_D|_{\lim}$ ($A\,m^{-2}$) attributed to the stirring effect in the reactor, its mean acoustic streaming and microjets generated by bubble collapses in cavitating media. Stirring in the neighborhood of the electrode can be expressed by the dimensionless Sherwood number [22–24]:

$$Sh = \frac{|\vec{j}_D|_{\lim} R_{\text{elec}}}{nDFC_{\text{sol}}},$$

$R_{\text{elec}} = 10^{-3}$ m is the electrode radius, $D_{Fe(CN)_6^{4-}} = (5.60 \pm 0.21) \cdot 10^{-10}\ m^2\,s^{-1}$ the diffusion coefficient of the species determined with a rotating electrode, F the Faraday constant, and C_{sol} the concentration of the species.

Experiments were carried out with a concentration in ferri-ferrocyanide $[Fe(CN)_6^{3-}] = [Fe(CN)_6^{4-}] = 5$ mM in a background salt NaOH = 0.2 M. We used a classical setup with 3 platinum electrodes; the working electrode can be located at various distances from the transducer. This limiting current density can be linked to an equivalent circulation of electrolyte U_{eq}, able to produce the same electrochemical signal in silent conditions, using equations of mass transfer and mass balance. Pollet and Hihn proposed the following equation [23]:

$$U_{eq} = \frac{1}{(0.45nFC_{\text{sol}})^2} D^{-4/3} \nu^{1/3} R_{\text{elec}} |\vec{j}_D|^2_{\lim},$$

where ν is the kinematic viscosity.

All experiments were conducted by a Tacussel PGZ 301 potensiostat.

3. Results and discussion

3.1. Velocity vector fields measured by PIV

The PIV method was previously used in many configurations with an interesting sensitivity [25,26]. Indeed the behavior of cavitation bubble field is dependent on many parameters such as acoustic power, reflector nature, i.e. whether the surface facing the transducer is free (case of a liquid without obstacle) or not (electrode surface) [4] and even on wave generation mode (continuous or pulsed). Therefore, first tests consisted in irradiating a free surface, whereas in the second test, an electrode was located in the acoustic field (giving an opportunity to compare with electrochemical measurements). It was observed that the polyamide particles (Rilsan 30 μm diameter) used as tracers formed cavitation germs, thus leading to a decrease in cavitation threshold (reach here for the lowest available powers instead of a few Watts in previous works [1]. The values of the axial velocity (fluid velocity on the acoustic axis) measured by PIV were averaged with a MATLAB post-treatment software, over a 5 × 5 mm^2 window, centered on the propagation axis, and sliding by steps from the vicinity of the transducer up to the free surface.

3.2. Irradiation of a free surface

The velocity vector fields obtained by PIV measurements for both HIFU transducers (Tfc750 and Tfc3000) for 20 W (acoustic power measured by calorimetry) are shown in Fig. 1. A conical shape is visible in both cases, with the same global pattern as the distribution of the active bubbles observed in a previous study [1]. Velocities reach their maximum values in the acoustic axis, and a whirlpool recirculating flow is visible close to the walls in the upper part of the reactor.

The average vectors are plotted as a function of transducer distance at various powers. Fig. 2a gives the results for the Tfc750 transducer. Velocities increase with distance and irrespective of power maximum values are reached of between 70 and 80 mm, slightly above the acoustic focal (90 mm), which corresponds to the maximum cavitation activity previously observed [1,27]. After the maximum, the presence of a large number of bubbles and the divergence of the acoustic field lead to fluid decelerations. Nevertheless, dependence of axial velocity on power is low, seemingly reaching a kind of saturation beyond 20 W and around 120 mm s^{-1}. On the contrary, an increase in axial velocity vs. power is noticeable in the zone close to the transducer. For example, if we look at its evolution at a given distance of 20 mm from the transducer, velocity evolves from a few mm s^{-1} at 10 W to more than 60 mm s^{-1} at 40 W. Results for the Tfc3000 transducer are shown in Fig. 2b. The same global trends are observed, i.e. the average value of the axial component of the velocity vectors increases as a function of distance to the horn up to a maximum of between 45 and 50 mm, but located just beyond the acoustic