CHARACTERIZATION OF THE ULTRASOUND ACTIVITY IN A LIQUID FLOW USING PARTICLES IMAGES VELOCIMETRY (PIV) AND ELECTROCHEMICAL MASS TRANSFER MEASUREMENTS

M. BARTHES\(^a\), G. MAZUE\(^b\), D. BONNET\(^a\), R. VIENNET\(^b\), J.Y. HIHN\(^b\), Y. BAILLY\(^a\)

\(^a\)Département ENERGIE, Institut FEMTO-ST, UMR CNRS 6174, Université de Franche-Comté, 2 avenue Jean Moulin, 90000 Belfort Cedex France.

\(^b\)Institut UTINAM UMR CNRS 6213, Université de Franche-Comté, 30 avenue de l’Observatoire, 25 009 Besançon, France.

\(^c\)Navy Clean, 21 Traverse Collet Redon, 13013 Marseille, France.

ABSTRACT: The present work is dedicated on the interactions between a liquid circulation and a perpendicular acoustic wave propagation. A specific experimental setup enables us to study one transducer operating at 20kHz, with the help of electrochemical mass transfer measurements combined with Particles Images Velocimetry (PIV) determination. Electrodes were located on the wall opposite to the acoustic emission. Experiments were performed for various Reynolds numbers: from 0 to 50 000 (different liquid flow rates and viscosities).

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 (Araújo et al., 2013), emulsification (Abismail et al., 1999), cleaning (Mazue et al., 2011). The main effect is already known to be the generation of cavitation bubbles, responsible for mass transfer enhancement, radical formation and interface cleaning. All those phenomena often contribute to an enhancement of the chemical reaction kinetics. Nevertheless, the dissemination of large scale functional unit in production is slow 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 actives zones. Some of them give a global quantification of the acoustic activity such as caloriometry (Romdhane, 1997), chemical radical dosimetry (Iida et al., 2005), whereas local information are given by determination of mass transfer (Trabelsi et al., 1996) or laser visualizations (Tomography (Mandroyan et al. 2009a) and Particles Image Velocimetry (Mandroyan et al. 2009b)). In papers available in the literature, acoustic activity is usually described in an initially motionless fluid bulk i.e. where the liquid motion comes only from the transducer activity (Trabelsi et al. 1996). Studies are then focused in the vicinity of the transducer horn, where the fluid flow is induced by the wave propagation (Mandroyan et al., 2010). Numerous large scale applications of ultrasound involved an additional liquid circulation inside the acoustic field. For example, the displacement of an acoustic cleaning tool at a surface in case of ship hulls cleaning (Mazue et al., 2011) or the circulation of reactants into a sonoelectroreactor (Gonzalez-Garcia et al., 2007). In this last case, electrodes are located face to 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: electrochemical measurements of mass transfer phenomena by cyclic voltametry using the well-known quasi-reversible redox couple Fe(CN)\(_6\)\(^3+\) / Fe(CN)\(_6\)\(^4+\), and Particle Image Velocimetry for velocity fields determination. PIV is a non-intrusive optical method which enables...
to measure 2D velocity vectors field in a plane of a flow (Raffel et al., 1998). Two successive laser pulses illuminate a plane of a particles seeded flow, so that a camera can record 2 consecutive images of particles in the plane. Then a Cross-correlation between these 2 images enables to calculate particles displacements during the delay time between the 2 recordings. Knowing this delay time and the spatial 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 (Pollet et al., 2007, Hihn et al., 2010), whereas mass transfer measurements allow a local evaluation of all stirring contribution at the electrode surface. It had been shown that the main contribution to the stirring at an electrode surface located face to the ultrasonic horn comes at 90% from asymmetric cavitation (Hihn et al, 2011). 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 50000) and equipped with transducers operating at 20kHz. PIV measurements show a great sensibility of velocity vectors fields induce by the transducer activity to the liquid flow rate increase, which can be quantified by electrochemical results.

MATERIAL AND METHODS

A dedicated set-up was especially design for those experiments. A cross-section polypropylene cell (40mmx40mm and 400mm length) of measure was included (figure 1). The transparent PMMA front window permitted the laser visualization. Controlled flow circulation was supplied by a centrifugal pump and the liquid was contained in the thermostated tank (25L). The ultrasound probe was embedded in a flange fixed at the cell bottom and electrode used for mass transfer determination was embedded in a flange fixed at the cell top. Three different locations of the electrode were possible.

For all experiments carried out at 20 kHz, a probe (Sonics & Materials, Danbury USA) was used as the ultrasound source. A titanium horn with a 25 mm radiating face was used to transmit ultrasound to the liquid media. The working electrode was a platinum disk (1mm 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 crown 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 measurement. The solution was potassium ferri/ferrocyanide (5.10^{-3} mol.L^{-1}) in sodium hydroxide (0.2 mol.L^{-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 sigmoidal current response yielding a signal plateau at mass transfer limited potential. The mass transfer limited current 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 U (Pollet et al., 2007) which represents global streaming at the electrode surface. In the specific case of a tangential flow to the electrode surface, \( V_p \) was calculated by means of the following equation (1) (Mazue et al., 2013):

\[
V_p = \frac{1}{(0.6 n F C_{\text{sol}} D^2)} \cdot D^{-4/3} \cdot v^{1/3} \cdot j_0^{-2/3} \cdot x^{1/3}
\]  

(1)

Where \( D \) is the diffusion coefficient (m\(^2\).s\(^{-1}\)), \( C_{\text{sol}} \) is the concentration of the electroactive species (mol.m\(^{-3}\)), \( v \) 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, \( j_0 \) is the current density (A.cm\(^{-2}\)) and \( x \), in this case (cf. Mazue et al., 2013), represents the distance between the electrode center and the beginning of limiting diffusion layer.

![Figure 2. Global field of the camera and location of the ROI and the electrodes.](image)

The experimental setup used for PIV measurements was composed of a dual beam pulsed Nd:YAG laser (532nm) paired with a camera (PIVCAM 13-8, 12bits). A synchronizer triggered 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 \( \Delta t \). During our experiment, this \( \Delta t \) was equal to 100ms. The liquid media was seeded with tracer particles (hollow glass beads, 8-12\( \mu \)m diameter). For each operating condition, a set of 100 pairs of images were recorded. For the image processing, we used the PYV software (Bonnet et al. 2011; Bonnet et al. 2012). First, images were pre-processed to correct the relative displacement due to the vibrations of the pump. Thus, all the images were corrected and had the same spatial origin. Then the Regions of Interest (ROI) were selected. For our study, we defined 4 ROI, in which vector fields and mean velocities were calculated. Three of them fitted a square of 8mm side length (256x256 pixels). These 3 ROI fitted the electrode location used for mass transfer measurement. The last ROI (1024x1056 pixels) represented the global field of the camera (Figures 1 and 2).
RESULTS AND DISCUSSION

PIV measurements allowed us to determine velocities vector fields on the observation window from which a spatial average velocity in each ROI was calculated. Those velocities are shown versus the Reynolds number of the liquid flow (Figure 3).

![Figure 3](image)

**Figure 3.** Spatial average velocities versus Reynolds numbers in the different ROI.

In the absence of liquid circulation (Re = 0), the activity in the center ROI (facing the transducer) was found to be higher than the ones in the upstream and downstream ROI. This result was already expected for a liquid flow induced by the transducer and directed toward the opposite wall (Figure 4.a). Then, this flow spread all over the impact area resulting in recirculating flows of equal magnitude, as shown by the upstream and downstream values (Figure 3).

![Figure 4](image)

**Figure 4.** Velocity vector fields at different Reynolds number

In silent conditions, spatial average velocities were proportional to the forced liquid motion and very similar to the velocities calculated from the ratio between flow and cross-section area. These results confirmed that spatial average velocities obtained from PIV measurements are relevant. When combining ultrasound and forced liquid flow, observations in the three local windows were different for the lowest Reynolds number (1800 to 3800). For both center and upstream windows, spatial average velocities decreased from Re=0 to Re≤3800. Then those velocities increased regularly, up to coinciding with the ones in silent conditions. Concerning the downstream window, the spatial average velocities increased with the addition of the forced liquid motion and reached a
maximum value for a Reynolds number of 2500. This observation can be explained by the
deveation of the convective flow plume to the downstream side. This also created a small
recirculation on its front, responsible of the velocities drop in upstream and center windows
previously described (Figure 4.b). For the higher forced liquid flows (i.e. for Re > 7000), the
convective flow plume was pushed out of the observation window as the influence of ultrasound
became negligible (Figure 4.c). The same behavior was observed while varying liquid viscosity
(from 10^{-3} to 2.25 \times 10^{-3}\text{Pa.s} – not shown here). The figure 5 shows the equivalent parietal velocity
(V_p) representing both tangential flow and the ultrasonic stirring from electrochemical
measurements. In the absence of liquid circulation, the V_p obtained for the three locations of the
electrode were less dispersed than spatial average velocities calculated in the three corresponding
ROI. This was particularly true for the central location, since the contribution of the stagnation
zone in the vicinity of the electrode was weakened in the PIV calculation area. In the downstream
measurement location, a maximum was also observed for 1800<Re<3800. This confirmed the
placement of the maximum of the agitation in the liquid circulation direction, even close to
wall. Furthermore, equivalent parietal velocities, determined in ultrasound presence, were always
larger than those obtained in silent conditions. Even for the highest Reynolds number (Re > 7000)
for which the convective flow plume was completely pushed out of the measurement window, a
part of the acoustic energy still reached the wall face to the transducer. This last result confirmed
that ultrasound remained effective at the wall opposite to the transducer, even for a high disturbed
hydrodynamic environment.

![Graph showing equivalent parietal velocity versus Reynolds numbers for different electrode locations](image)

**Figure 5.** Equivalent parietal velocity versus Reynolds numbers for different electrode locations

**CONCLUSION**

In the literature, there is a lack of available references concerning the characterization of an
acoustic field disturbed by a liquid flow. Both Particle Image Velocimetry and electrochemical
methods used in the present work appeared to be relevant and bring complementary information.
The most interesting result showed that the ultrasound contribution remained noticeable on the
surface opposite to the transducer even in the presence of a liquid circulation perpendicular to the
wave propagation. The full description of fluid motion dynamics remains complex and is currently
investigated.
REFERENCES