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## Reassessment on the Operation of a T-mixer Chemical Reactor in the Cavitation Regime

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In a previous work in our team (Oualha et al., Chem Eng Trans, vol 73, 67-72 (2019)) it was shown that at large Reynold numbers ( $Re > 8000$ ) a vapor phase appears by cavitation in T-mixer reactors used for chemical precipitation of nanoparticles. This phenomenon can affect dramatically the properties of the obtained nanoparticles. In the work of Oualha et al (2019), the apparent size (more precisely the hydrodynamic diameters) of vapor bubbles due to the cavitation, was in situ monitored in the reactor by using SLS (Static Light Scattering) and DLS (Dynamic Light Scattering) methods. A modeling was also given using Fluent software: calculated size population of bubble was provided. Unfortunately this population could not be directly compared to experimental data since the relationship between apparent and real size was unknown. In the present work, new DLS measurements using particles of calibrated sizes are presented to help us to interpret and comment our previous results on bubbles size distribution. They were obtained using a new apparatus designed and realized for the purpose of defining a method of true diameters measurement.

Keywords: T-mixer, Cavitation, Light scattering (DLS/SLS).

### 1. Introduction

The reactor T-mixer depicted in Figure 1a has been used by our team to synthesize nanoparticles by sol-gel process.

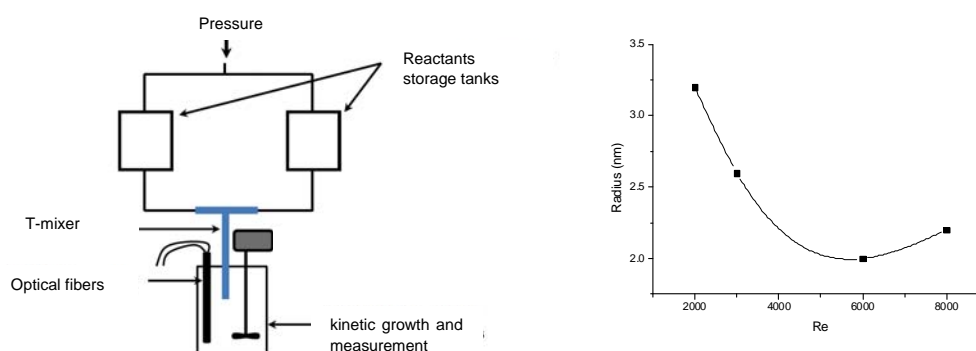


Figure 1: Simplified scheme of sol-gel process used in our group for preparation of nanoparticles (a) and mean radius of  $TiO_2$  nanoparticle obtained with the sol-gel process at various Reynolds (b) [Oualha et al., 2017]

Various nanoparticles of  $TiO_2$  (Rivalin et al., 2003),  $ZrO_2$  (Labidi et al., 2015), Ag (Jia et al., 2012) etc... were prepared. It has been observed that the mean radius of the nanoparticle depends on the injection conditions

(pressure and velocity) of reactants. In the case of  $\text{TiO}_2$  the plot of mean radius vs Reynolds number ( $\text{Re} = \rho \cdot u \cdot D / \mu$ ) exhibits a minimum (see Figure 1b). Oualha et al. (2017) showed that this minimum is related to the appearance of a vapour phase by cavitation. With pure water, cavitation appears in T-mixer reactor for  $\text{Re} > 8000$ . This phenomenon occurs when the local pressure of a liquid is reduced under the saturation pressure. To control the process of nanoparticle precipitation in the T-mixer, it is necessary to know and control the sizes of vapor bubble generated by cavitation. In a previous work (Oualha et al., 2019) such study was carried out using dynamic light scattering (DLS). See Figure 2 below obtained by Oual ah et al (2019).

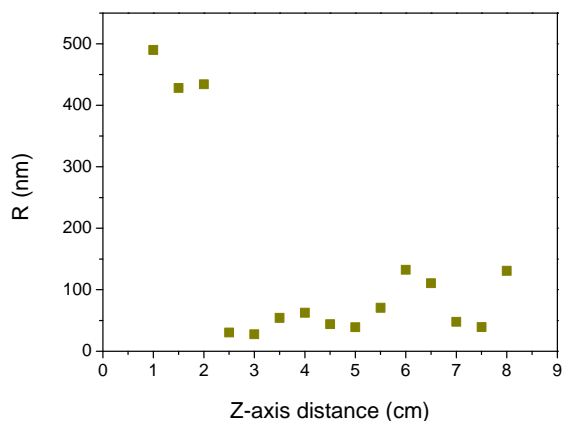


Figure 2: Mean radius of cavitation bubbles along z-axis of outlet tube of T-mixer [Oualha et al., 2019]

However this method allows measuring hydrodynamic diameters but not directly the true diameters. Additional study was thus needed to find the relation between true and hydrodynamic diameters. This is the goal pursued in the present paper.

The present study completes then the previous one. A brand new apparatus has been designed and realized for that purpose. The dynamic light scattering (DLS) measurements were carried out using latex beads of known size. Three different sizes were investigated in the flow range  $500 < \text{Re} < 9000$ .

## 2. Experimental Setup

To characterize the particle sizes, we have conceived and built a new experimental device. The scheme is depicted in Figure 3.

The principle is as follows. An aqueous solution of latex beads (see next section for more characteristics) is circulated in a straight transparent glass tube of 4 mm internal diameter allowing SLS-DLS (static and dynamic light scattering) optical measurements. Unlike the work of Oualha et al. (2019), operating conditions are without cavitation so that the flowing suspension is composed only of solid particle in water liquid solvent.

The Latex beads used in this work were provided by Sigma-Aldrich. Three different radiuses were investigated: 0.55  $\mu\text{m}$ , 1.5  $\mu\text{m}$  and 12.5  $\mu\text{m}$ . The original solutions before dilution were mainly composed of polystyrene (30%) and water (69 %).

To ensure a flow range between 0.1 and 1.66 L/min, we used two pumps: one is peristaltic, provided by Donko and allows to work between 0.1 and 0.57 L/min, the other one, provided by Verder (hydra -cell G13), ensures a high flow rate from 0.67 L/min up to 9.3 L/min.

The light scattering measurements were performed following the procedure described by Oualha et al. (2019). A single frequency 40 mW / 640 nm Cube 640-40 Circular (Coherent) laser was used with two monomodal optical fibers placed very closely to the tube surface (and 3 mm away from the tube center). One fiber emits and transmits the laser light to the tube and the other one receives the scattered photons at a  $90^\circ$  angle from the incident light. The analysis of the scattered light was carried out using a digital 255 channels photon correlator (PhotoCor Instruments). The measurements were performed in automatic sampling mode with a data storage period of 60s.

All experiments were conducted at temperature of 20  $^\circ\text{C}$ .

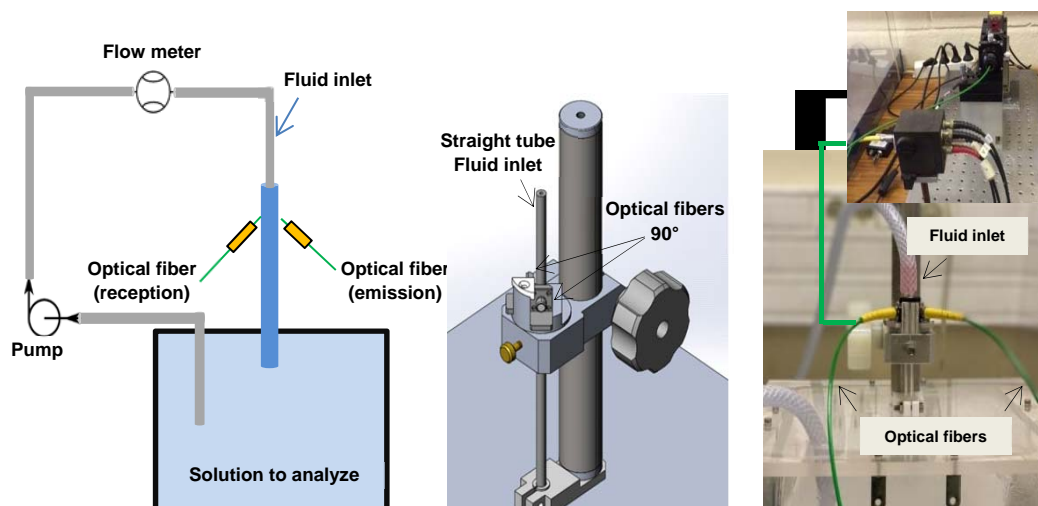


Figure 3: Scheme of the experimental device

### 3. Results and discussion

Auto-Correlation Functions (ACF) obtained by DLS are widely used for determination of particle diameters. In most cases this is done in a solution at rest and the procedure is well known. However, if the solution is flowing, the hydrodynamics appears to have a strong impact on the ACF even in the laminar regime as shown in earlier studies (Leung et al., 2006). This impact is not very well quantified but has to be taken into account in diameter determination. Here are presented our first measurements made to gain a better understanding of this phenomenon.

In that purpose, three series of ACF were acquired for the particle radii investigated here:  $0.55\ \mu\text{m}$ ,  $1.5\ \mu\text{m}$  and  $12.5\ \mu\text{m}$  in a large range of flow rate from  $0.1\ \text{L/min}$  to  $1.66\ \text{L/min}$ . In those conditions the Reynolds number ranges from 500 to 9000.

The volumetric fractions of the different latex beads with respect to water solvent were set to values reported in Table 1 by dilution of the original solutions (see section above). This dilution realizes a good compromise between signal intensity and the necessity of reducing of multiple scattering (increasing also with concentration).

Table 1: Volumetric fractions of latex beads solutions used in DLS measurements.

Particles radius ( $\mu\text{m}$ )	Volumetric fractions
0.55	$2.17 \cdot 10^{-4}$
1.5	$8.6 \cdot 10^{-3}$
12.5	$3.44 \cdot 10^{-4}$

Examples of such ACFs for latex beads of radius  $R = 0.55\ \mu\text{m}$  are given on Figure 4.

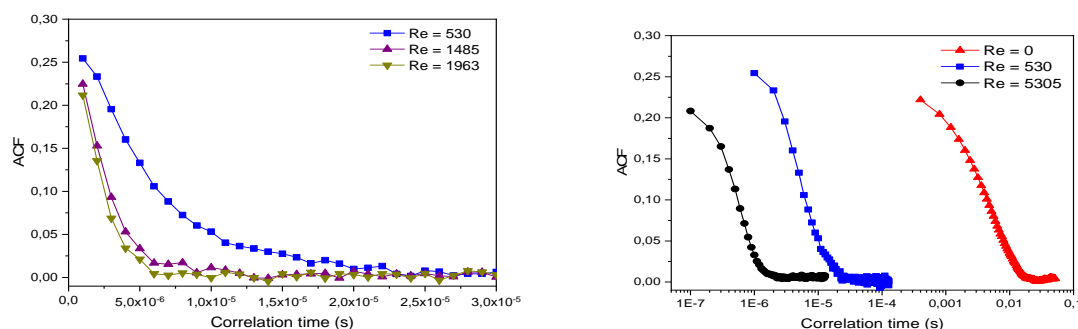


Figure 4: Auto Correlation Function (ACF) of Latex beads  $R = 0.55\ \mu\text{m}$  at different Reynolds

This figure clearly illustrates how important is the fluid hydrodynamics on the ACF curves. As a general feature, the ACF value at a specific correlation time decreases considerably when Reynolds number increases.

Such a behavior has already been observed in the past in a pipe in the laminar regime at low Reynolds (see Leung et al., 2006: Re up to 1630) and also in the turbulent regime by several authors (Chowdhury et al., 1984 up to Re = 3380 in a pipe; Pak et al., 1991 Re = 375 -1490 in a plenum chamber followed by a grid and a water tunnel). A careful examination of the literature on the subject, shows that the data are rare, scarce and measured at moderate Reynolds values far below the ones at which our T-mixer operates. Clearly there is a lack of systematic experimental measurements that has to be filled in and this is the purpose of this work.

In order to have an efficient method of diameter determination (exploitation of ACFs), the effect of hydrodynamics on the ACF has to be well understood from a theoretical point of view. In such a goal, let us recall that an experimental ACF is generally represented by:

$$g^{(2)}(\tau) = A + B e^{-2G\tau} \quad (1)$$

Where A is the baseline, B is an empirical experimental constant, and  $\tau$  is the time. G is the decay constant, which in the case of a fluid at rest is linked to the Brownian motion by:

$$G = -Dq^2 \quad (2)$$

D is the Brownian diffusion coefficient and q is the magnitude of the scattering wave vector. The radius R is related to D by Stokes-Einstein equation:

$$D = \frac{kT}{6\pi\mu R} \quad (3)$$

Where  $k = 1.38 \cdot 10^{-23} \text{ J K}^{-1}$  is the Boltzmann's constant and T is the absolute temperature.

Equations (1) and (2) strictly apply to fluids at rest. In the case of flowing fluids, one has to take count of velocity gradient  $\dot{\gamma}$  and / or mean velocity u. Eq(1) becomes then:

$$g^{(2)}(\tau) = A + B e^{-2G_{app}\tau} \quad (4)$$

where  $G_{app}$  (app for 'apparent') can be written:

$$G_{app} = -Dq^2 + F(\dot{\gamma}, u, \tau) \quad (5)$$

F is a function depending strongly on the flow regime (and the flow field). A few expressions have been given in the case of simplified assumptions and geometry. Maloy et al. (1992) provided a general and formal treatment in term of probability density of velocity differences. Ackerson and Clark (1981) supposed a constant shear flow. Fuller and Rallison (1980) considered a cylindrical geometry. Nijman (2002) summed up most of the expressions in laminar and turbulent regimes. However, no general expression appears widely accepted especially in the turbulent regime (Goldburg, 1991). Finding a theoretical expression is all the more difficult that we deal with suspensions of particles. The transport of particles in a liquid flow is a complex subject, and the velocities of particles (potentially different from the fluid velocity) may depend dramatically on their size and their interaction with eddies in the turbulent regime. As shown by figure 5, the function F become predominant (several decades of difference) over  $-Dq^2$  term in the turbulent regime as well in laminar regime of high Reynolds. Around 3-4 orders of magnitude are observed between zero and high velocities conditions. This effect is particularly pronounced for the small particles that are more easily transported than the big ones by the flowing fluid. Also other characteristics of the flow field (shear Reynolds stress profiles, correlations strength of turbulent motions, see Shokri, 2017) may depend strongly on size. All the pertinent parameters will have to be identified and their effect will have to be clarified in future studies.

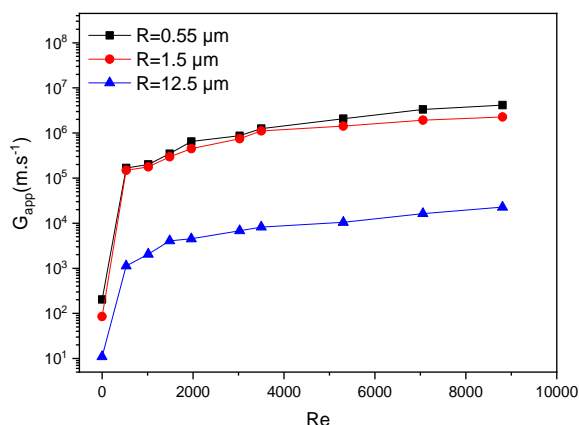


Figure 5: Apparent decay constant  $G_{app}$  for three size of latex beads (0.55, 1.5 and 12.5  $\mu\text{m}$ ) with Reynolds

It appears this function  $F$  could not be ignored in the diameter measurement. Supposing eq(1) and eq(2) are true would lead to an apparent hydrodynamic radius much smaller than the true radius as emphasized by the figure 6. From our measurements, the apparent diameter varies quickly with  $Re$  in the laminar regime, and seems to stabilize to a constant value at high  $Re$  turbulent regime.

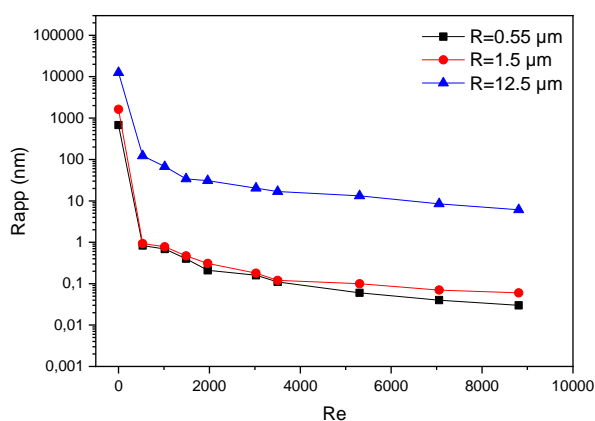


Figure 6: Apparent radius of particle with Reynolds number. Uncertainty on the apparent diameter is estimated around 20 % (based on average values of 10 ACFs).

At the present status of this work more data acquisitions have to be made especially with particles of other radius in order to obtain quantitative correlations (not enough data at this state). Also it is may be necessary to cover an even larger range of diameter and Reynolds number.

Also the applicability of these results to bubbles in the T-mixture deserves a comment. It appeared clearly from the discussion above that the ACF and thus apparent diameters depend on the flow field and its impact on particle velocities and their ability to follow or not the liquid flow. However, the ability of following the flow depends on the mass of the particles and thus on their densities. Bubbles have lower densities than solid particles and are expected then to flow more easily. This has to be quantified to a theory, probably through additional measurements.

#### 4. Conclusions

In the present status of the study it is not strictly speaking possible to determine the true value of the bubble diameter. But taking count that there is several order of magnitude between true and apparent radius in the high turbulent regime (at least for solid particles), one can infer that the true bubble radius is probably at least of order of several  $\mu\text{m}$  or tenth of  $\mu\text{m}$ . Such a high difference between true and apparent diameters was unexpected. This remains of course to be confirmed on a firmer ground and quantified. Model and experiments are expected to play complementary roles in this perspective.

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