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Particle deposit formation during filtration characterisation using ultrasonic waves

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Membranes filtration processes are widely used because of their ability to remove particles, colloidal species and micro-organisms from different liquids feeds. However an inherent process limitation is the membrane fouling due to deposition of suspended matter during filtration. Therefore the understanding of formation and transport properties of particle deposit responsible for membrane fouling is a necessary step to optimize membrane processes. These deposits are non homogeneous, highly porous and very thin (less than 500 µm).Thus, it is necessary to obtain local information in order to analyze and model the basic mechanisms involved in deposit formation and then to further predict process operation. As local parameters such as cake thickness and porosity are hardly reachable with conventional techniques, we propose in this paper the use of an ultrasonic echographic method. In a first step this method is validated on deposits of small glass balls. We show that the porosity and thickness of the deposit is in good agreement with theory. Then, the ultrasonic technique has been adapted on a filtration cell in order to give in line results during filtration. First results are presented and discussed.

1 Introduction

In order to study deposit formation, ultrasonic methods based on normal incidence reflectometry are generally used in literature and are based on a very simple principle: an ultrasonic signal propagates from an ultrasonic transducer to the membrane, is reflected on the membrane and is received by the transducer.

If a deposit is present on the membrane, the ultrasonic signal is reflected on the deposit and consequently its propagation time is smaller. If this reduction of propagation time is measured, the thickness of the deposit can be evaluated. One can also study the amplitude of the ultrasonic signal in order to obtain results on reflection coefficient, density, roughness…

In literature, Mairal et al. [1] were the first to use ultrasonic reflectometry for the observation of membrane fouling. They monitored in situ fouling of inorganics ($CaSO_4$) in a cross-flow reverse-osmosis system with a flat-sheet module. The relative amplitude of the ultrasonic amplitude was followed in conjunction with flux decline. Ultrasonic signal measurement provided sensitivity to the dynamics of the fouling layer growth that was comparable to that observed from the flux decline behaviour. In this study ultrasonic measurement could only provide qualitative information about the fouling layer.

Li and al. [2] also used ultrasonic reflectometry for the study of fouling on flat-sheet nylon membranes during cross-flow microfiltration at 100 kPa. The feed solution was an effluent from a wastewater treatment plant. The acoustic signal had a frequency of 10 MHz. Results obtained from this study suggest that the combination of flux determination and ultrasonic measurement can provide a much clearer view of the fouling behaviour of a membrane than flux decline alone that is also sensitive to charge accumulation and membrane compaction. Once fouling was initiated, the acoustic impedance difference and topographical characteristics at the feed solution/membrane interface will change resulting in a new echo. With this new echo, it was possible to quantify the fouling layer by calculating its thickness. Besides, the fouling echo amplitude is an indication on the state of the fouling layer. The dense the fouling layer is, the better the reflection is (and thus the larger is the amplitude that is seen). In this study it was possible to obtain quantitative information.

So, from literature it is clear that ultrasonic methods are able to give information about the fouling and the deposit growth.

As we will see further, in our case, the involved thicknesses are very small (< 500 µm) and high ultrasonic frequencies are highly attenuated. So, one has to work with relatively small frequencies around 5 MHz) on small thicknesses. Consequently various echoes reflected from the system (membrane + deposit) interfere leading to complex ultrasonic responses. Such responses are also obtained by Li et al. Regarding their signals it is clear that extracting precise information is quite difficult and hazardous.

In oder to overcome such a difficulty, we propose now a specific acquisition mode and signal processing. After the presentation of this specific signal acquisition and processing used, in order to separate the signals, a first validation on glass spheres deposits will be presented. Then, first “in line” results on a real filtration cell will be given and discussed.

2 Normal incidence reflectometry

2.1 Signal acquisition and processing

Ultrasonic longitudinal 5 MHz waves generated with a Panametrics plane transducer first travel into the solution to be filtered and are reflected on the membrane.

If there is no deposit on the membrane, if “L” is the distance between the transducer and the membrane and if V_s is the ultrasonic longitudinal velocity in the solution, the arrival time of the echo is $2L/V_s$.

Now, if a deposit of thickness “e” appears, a new echo reflected on the top of the deposit arrives for the time: 2(L-e)/Vs. Consequently if V_s is known and if the difference of arrival time Δt between the echo at the beginning of filtration and the echo for a time t of filtration is measured, the thickness “e” can be measured.

We will see further that the solutions to be filtered are made of water and clay with small concentrations. In these solutions, V_s has been measured and can be considered equal to the longitudinal velocity in water: 1500 $m.s^{-1}$.

On a practical point of view, echoes are recorded for regular time intervals during the filtration, encoded in colours and store in an image, line after line. So, at the end of the filtration one has a (time x time) picture representing the various echoes acquired during filtration.

As during filtration, the thickness of the deposit grows, the echo reflected on the deposit surface arrives earlier and earlier. If the velocity in the deposit is not too different from the velocity in the solution to be filtered, the signal reflected on the membrane does not move a lot. (cf Fig 1. and Fig 2.)

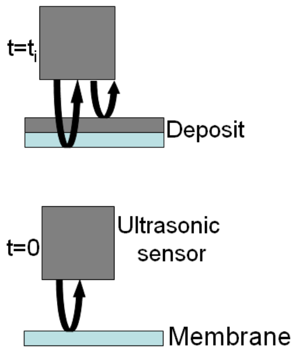

Fig 1. Ultrasonic travels considered during the filtration

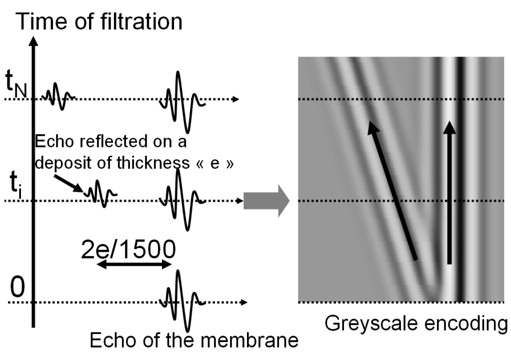

Fig 2. Data acquisition during the filtration

At the beginning of the filtration, the deposit is so small that echoes are overlapping. But, as the echo reflected on the membrane does not evolve a lot, we subtract the echo for t=0 to all the other traces. Hence we obtain a picture on which only the echo reflected on the top of the deposit is present. One can also use specific algorithm in order to separate the waves such as KLT. Details about this algorithm can be found in references [3][4][5].

Then the thickness can be deduced from the beginning of filtration. Concerning the time of flight evaluation an intercorrelation procedure has been implemented on Labview software.

2.2 Validation on glass spheres

In order to validate our data acquisition and processing, tests have been performed for glass spheres sedimentation. For these experiments the following sedimentation cell has been built (cf Fig 3). In order to check the thickness measured using ultrasonic waves, a web cam has been used to visualize the deposit formation. Such a procedure can be used here but is not applicable on the real filtration cell which will be presented in part 3. Furthermore, in industrial processes, if cells are not transparent, only ultrasonic signal can be used.

In figure 4 we present an example of picture acquired and thickness measured with ultrasonic method and web cam. The good agreement between the two methods validates the ultrasonic approach.

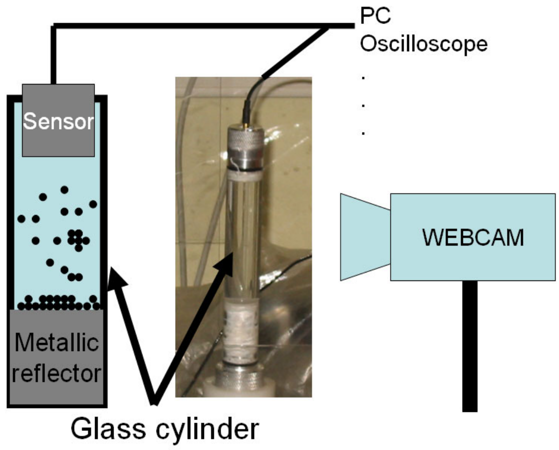

Fig 3. Sedimentation cell built

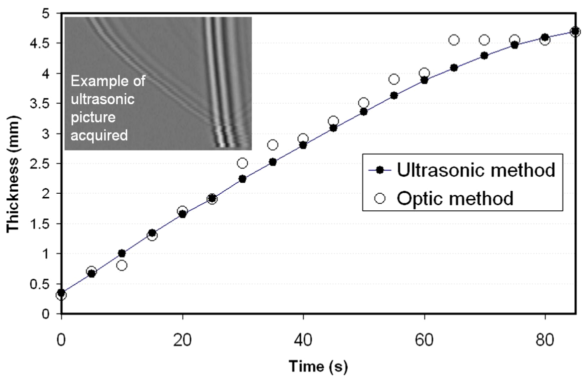

Fig 4. Validation of the ultrasonic method for sedimentation of 150 µm glass spheres

At last, various tests have been performed for glass spheres having the following diameters: 22± 2 µm, 70± 20 µm and 100 ± 20 µm. For each test, a mass m=0.5 g of spheres have been put in the sedimentation cell.

For glass spheres falling in water, it can be consider that only the Stocke’s force due to water viscosity acts [6][7]. Using this force and the Archimedes one, it can be demonstrated that balls fall with a constant velocity after less than 1s of falling (transitory regime). This constant velocity leads to a linear evolution of the deposit formation versus time. When the sedimentation is finished leading to a thickness “e”, we can write :

$$c = \frac{m}{\rho \pi R^2 e} \qquad (1)$$

Where c in the compactedness (it means 1-p where p is the porosity), ρ is the density of glass, R the radius of the sedimentation cell.

So the compactedness of the layer can be deduced. It should not depend on the spheres sizes. On the following graphs thickness evolution for various sizes of spheres, compactedness deduced and minimum thickness obtained versus the sphere sizes are presented.

One can see that the linear behaviour is observed at the beginning of the sedimentation.

We are able to detect quite a monolayer of spheres. This demonstrates that the echoes deconvolution is efficient for very small thicknesses.

At last the compactedness measured seems not to depend on sphere sizes as predicted.

Furthermore the mean value obtained (around 57 %) is in good agreement with what is generally observed for granular media : compactedness between the Random Loose Packing (65 %) and the Random Close Packing (55 %) [8].

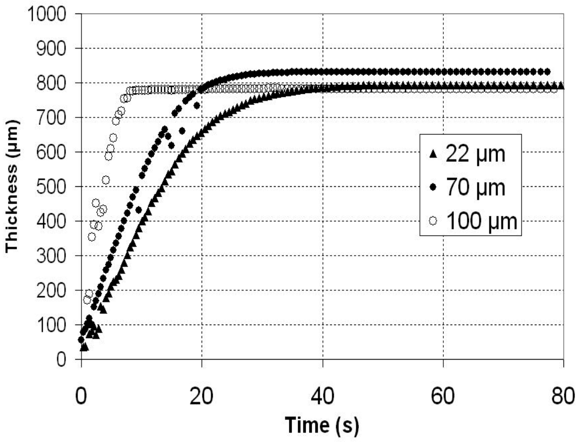

Fig 5. Thickness measured versus time for the sedimentation of glass spheres of various diameters

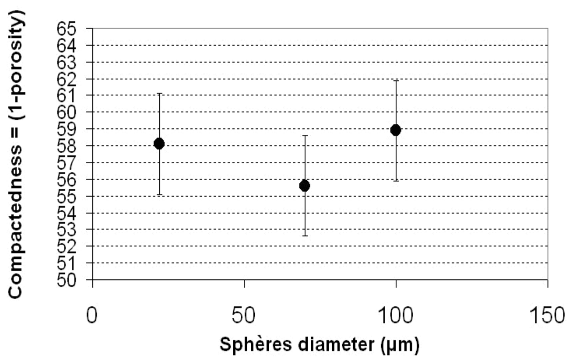

Fig 6. Compactedness measured for each sphere size

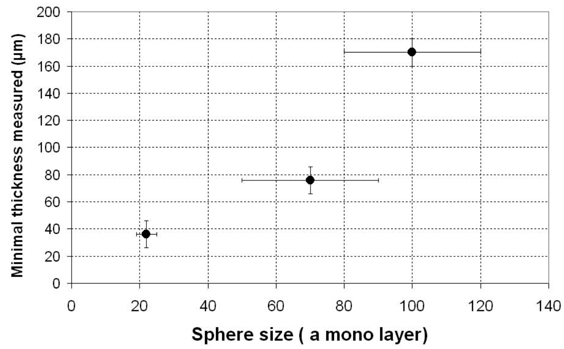

Fig 7. Minimal thickness measured versus sphere size

3 First results on the filtration cell

3.1 Filtration cell description

A narrow rectangular channel with one membrane wall was designed. This geometry introduces some confinement effects similar to those occurring in an inside/out hollow-fiber and allows to observe their influence on the deposit build-up.

Each of the two parallel plates of a flat transparent Plexiglas chamber was machined with a 28.2 cm height and 4 mm large and depth channel. A flat-sheet membrane (Polysulfone, Molecular weight cut off (MWCO=100 kDa)) was put between the two plates; thus filtration is operated only on one channel wall (112.8 10^{-5} m^2 of effective filtration area). This filtration device is operated in dead-end at constant transmembrane pressure (TMP). Liquid feed can be introduced from the bottom or from the top of the channel. Each of the two plates was connected to a piping network that enables to operate filtration.

The filtration cell and the ultrasonic sensor can be visualized on the following figure :

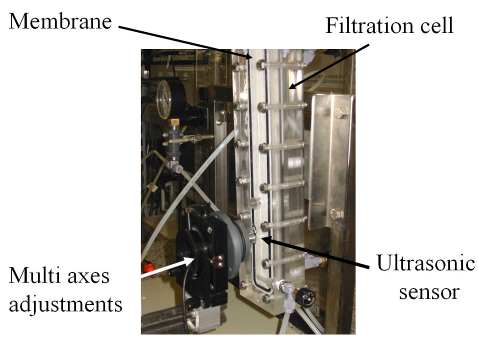

Fig 8. Experimental device for in line measurements

3.2 First experimental results

Clay suspensions were used so as to model ground water. Clay particles were bentonite particles with a mean diameter of 5-6 µm. Suspension concentration 1.5 $g.l^{-1}$. The thickness measured versus time is presented in figure 9.

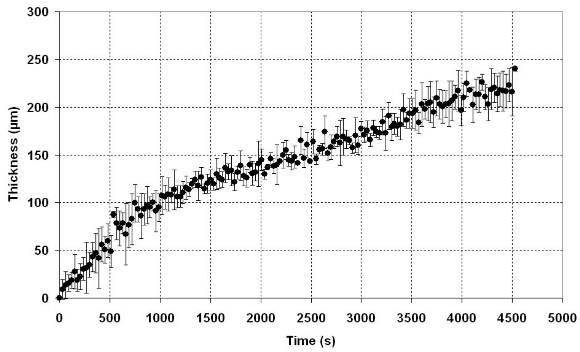

Fig 9. Thickness measured during a real in line filtration

3.3 Discussion

Using an optical method designed and built in the INSA of Toulouse, and based on the deviation of a laser sheet [9] on the deposit it is also possible to evaluate the deposit growth. First tests performed with this optical method and compared with the acoustic approach seem to show that the thicknesses evaluated with the two methods would not be totally equal. Indeed it seems that the optical thickness is smaller than the acoustic one. But the velocities of deposit growth seem to be equal. These preliminary results obtained in PhD thesis reference [10] have to be checked. This difference could be due to the fact that during the filtration the interface is not sharp at all but is constituted buy a gradient of porosity. In this case, the interaction of optical or acoustical is not as simple as for sharp interface and further works have to be performed.

4 Conclusion an perspectives

As mentioned in literature we have demonstrated in this paper that ultrasonic high frequency waves are very efficient to evaluated deposit growth during filtration on filtration cells in which optical methods are not easily applicable. Results obtained have been validated for sharp interfaces and have now to be refined for interfaces made of porosity gradients. Furthermore, other information such as ultrasonic velocity and attenuation in the deposit are going to be evaluated an studied.

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