EXPERIMENTAL STUDY ON THE ULTRASOUND-INDUCED BEHAVIOUR OF GAS POCKETS ENTRAPPED AT A SOLID/LIQUID INTERFACE

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ABSTRACT:
In this paper, the ultrasound-induced behaviour of gas pockets entrapped at a solid/liquid interface is experimentally studied. Indeed, this kind of interface can induce a significant decrease of the transmitted ultrasonic energy and therefore can reduce the performance of ultrasonic inspection in sodium cooled fast reactors for instance. To explain this event, a hypothesis has been formulated: under the effect of ultrasound, the gas pockets could coalesce and prevent the passage of the ultrasound. This hypothesis can be studied by visual observation of the phenomenon. This paper is presenting an original experiment which simulates this phenomenon in water. This experiment consists in observing gas pockets subjected to an ultrasonic field over various time scales. The results show that coalescence does not occur. It is concluded that real conditions are not favourable to the occurrence of the coalescence phenomenon. According to these conclusions, another theory is envisaged.

KEY WORDS: ultrasound, wetting, coalescence, roughness, sodium-cooled fast reactors.

INTRODUCTION:

The non-destructive testing by ultrasound of the structures of a sodium-cooled fast reactor (sodium is a liquid metal) must overcome the difficulty to transmit signals to the steel/sodium interface.

If the solid/liquid system is non-wetting and if the solid surface is rough enough so it is likely that microscopic gas pockets be trapped between the liquid and the solid [1,2]. Such an interface (with the simultaneous presence of the solid, liquid and gas phases) is named composite. This composite interface induces a significant decrease of the transmitted ultrasonic energy. To explain this phenomenon, a hypothesis was formulated: under the effect of ultrasound, the gas pockets could merge, forming a gas film which would prevent the passage of the ultrasound.

An equivalent experiment with water [3] was conducted to avoid the constraints related to the use of sodium. Indeed, in water, the coalescence hypothesis can be studied by visual observation of the phenomenon. This is the purpose of an original experiment described in this document. The experiment consists in observing the ultrasound-induced behaviour of gas pockets.

In this paper, first, the experimental device is described then the coalescence hypothesis is defined. The results of two separate studies are next presented: the first consisting in observing the ultrasound-induced behaviour of gas pockets on a long time scale (some tens of minutes), and the second on a very short time scale (a few microseconds). In each study the results are analysed and considering the observation of another phenomenon provides an interpretation.

EXPERIMENTAL PROCEDURES:

It is first a question of creating artificially, in water, composite interfaces whose gas surface fraction is controlled and represents a parameter of study. For that, eight silicon substrates are used: one control substrate (smooth, no grooves) and seven engraved substrates. The substrates used are wafers which are very smooth (Ra < 1 nm) on both sides and engraved on one side only. The engraved side is a model rough surface. This controlled roughness takes the form of a regular array of parallel rectangular grooves (Fig. 1). The grooves have a width of 20 µm (2r) and depth of 30 µm, separated by the distance (e) which is different for each substrate. The surface fraction of purely solid/liquid contact, τ, is defined as the ratio of the solid/liquid contact area divided by the apparent area of the substrate. For each substrate, the value of τ is calculated considering a composite interface: liquid/gas at the grooves and solid/liquid between the grooves. For the control substrate τ = 1.

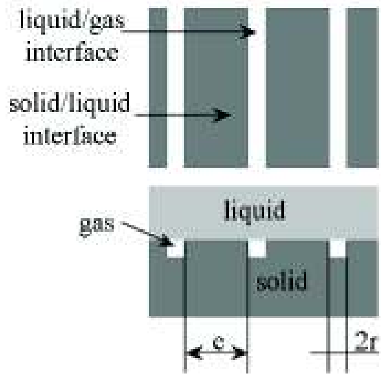

Fig. 1: Diagrammatic representation showing the front (left) and the section (bottom) of the engraved face of a substrate immersed in water.

To obtain a composite interface, the surface of the substrates is given chemical treatment to make it hydrophobic. A very thin layer of octadecyltrichlorosilane is grafted onto the surface of the substrates by a process used in liquid phase [4,5]. When the substrate is immersed in water (Fig.1), the water cannot enter the grooves due to the surface hydrophobia and the narrow width of the grooves.

The experimental setup consisting of the following four assemblies: mechanical assembly, ultrasonic assembly, optical assembly and image acquisition chain, is represented diagrammatically on Figure 2. The substrate is immersed in water at ambient temperature: about 18 °C. It is subjected to an ultrasonic field with the engraved surface in front of a transmitter transducer and at normal incidence. A transducer identical to the transmitter records the ultrasonic signal transmitted across the substrate. The wave trains imposed are of 10 periods of central frequency 1 MHz (i.e. a signal duration of about 10 µs). This frequency lies within the range of frequencies typically used for ultrasonic inspection.

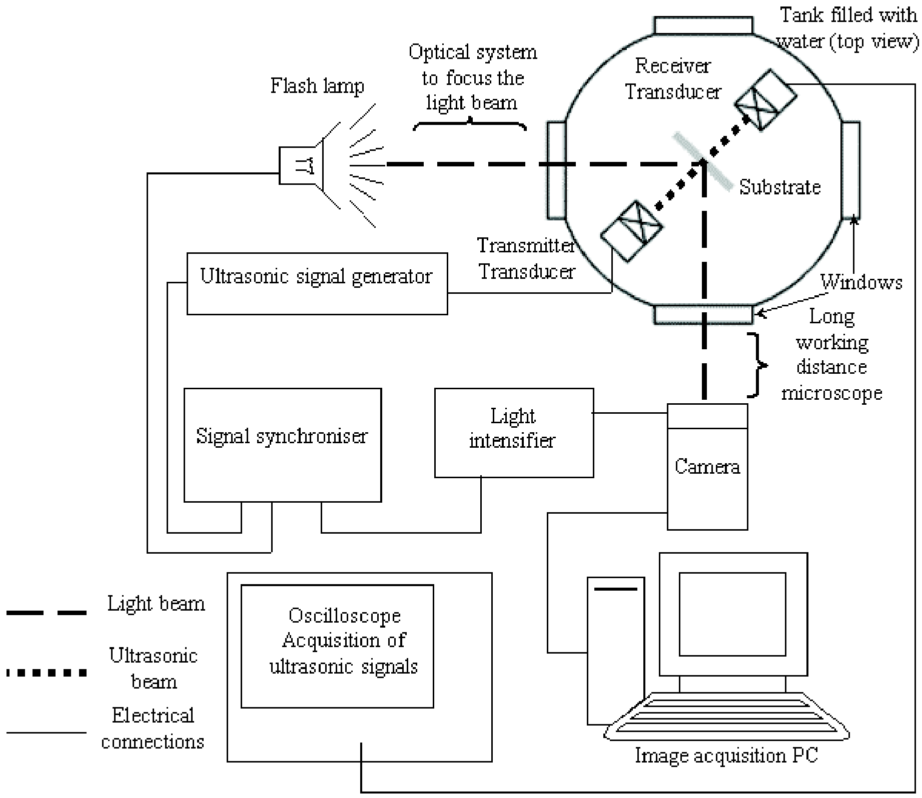

Fig. 2: Diagram of the experimental setup.

The experiment consists in observing a small area of the substrate surface. Photographs and videos are recorded with a camera through a long working distance microscope to obtain a total magnification (optical + numerical) of 386, producing an image measuring 12×9 cm. Movement blur is minimised by choosing an exposure time of 200 ns obtained using a flash lamp and a light intensifier.

On the images obtained (Fig. 3), the grooves filled with water are dark across the entire width of the groove whereas those filled with gas have a variable brightness profile across the width of the groove: they are light at the centre and dark at the edges. In both cases, groove filled with water or gas, the light beam which enters the groove (beam refracted by the liquid/gas interface for the case of the groove filled with gas) is assumed to be "trapped" in the groove. However, when the groove is filled with gas, some of the beam is reflected by the liquid/gas interface and projected onto the camera lens, but its brightness is less than that of the incident beam. Moreover, if the interface is curved it acts like a lens, converging the reflected beam. This explains the variable brightness profile across the width of the groove between the centre and the edges.

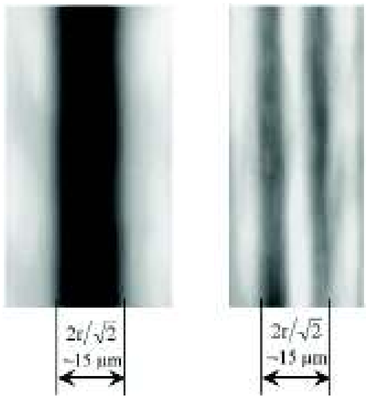

Fig. 3: Left: vertical groove filled with water. Right: vertical groove filled with gas.

ORIGIN AND DEFINITION OF THE COALESCENCE HYPOTHESIS:

The comparison between the ultrasonic transmission through a completely solid/liquid interface (control substrate) and through a composite interface is characterized by the interface transfer function in transmission mode ITF_t:

$$ITF\_t = \frac{A(f)}{A_{Ref}(f)} \qquad (1)$$

where A(f) is the modulus Fourier transform amplitude of the ultrasonic signal transmitted at normal incidence across the studied substrate and Aref(f) that transmitted across the control substrate acting as reference. The amplitude is measured at the frequency f equal to 1 MHz.

The results obtained with substrates S1, S2, S3, S7 and the control substrate demonstrate a significant drop in ITF_t when τ becomes less than about 90 % (Table 1). This drop is much greater than the simple decrease induced by the reflecting surface formed by the gas pockets. To explain this phenomenon, Lesueur [3] suggested that the ultrasound led to a dilatation of the gas pockets, then to their coalescence. A second important phenomenon observed is the slow increase of the ultrasound signal amplitude. It is assumed that it is due to the fact that the proportion of gas at the interface decreases: the initially composite interface gradually becomes a purely solid/liquid interface.

substrate	control substrate	S1	S2	S3	S7
τ	1	0,9	0,82	0,72	0,36
ITF_t	1	0,49	0,19	0,09	0,01

Table 1: Values of ITF_t for different values of τ (each value of τ corresponding to one substrate) at 1MHz.

The purpose of this experiment is to observe the interface and the shape of the gas pockets before and after their exposure to an ultrasonic field. The aim is to observe whether this ultrasonic field induces an enlargement of the gas pocket then their coalescence, i.e. the formation of a gas film. The bibliographic study allows making certain hypotheses regarding the behaviour of the gas pockets in the grooves subjected to an ultrasonic field. During one period of the ultrasonic wave, they will undergo alternately one compression phase and one expansion phase. Two possible types of behaviour which could result in coalescence are considered.

In the first case, the gas pocket interface oscillates during the passage of the ultrasonic wave. The volume of the gas pockets therefore fluctuates around a mean value. It increases, however, during the successive ultrasonic cycles by rectified diffusion [6] until coalescence occurs. In this case, it is difficult to predict the time interval to obtain coalescence. This time interval depends on the duration of the ultrasonic waves (a period or a wave train) and on the wave repetition frequency. For example, if the transducer transmits pulses at a very low repetition frequency, coalescence may never occur.

In the second case, if the acoustic pressure amplitude is sufficiently large, as soon as the first ultrasonic wave expansion cycle occurs, the gas pockets expand to such an extent that coalescence is obtained immediately. In this first case, coalescence is obtained within a very short time interval: of the order of the ultrasonic period, i.e. 1 µs or less. In an experiment quite similar to that described in this document, Bremond et al. [7] use a lithotripter which generates depressions of a few MPa. They manage to expand gas pockets which were initially completely contained in cylindrical holes of diameter 8 µm. This expansion results in the formation of a hemispherical cap of gas above the cylindrical hole, of diameter much greater than that of the hole since it can reach up to 400 µm. Neglecting the effect of the wall (solid silicon surface) and assuming the analogy with half of a free bubble, Bremond et al. use the Rayleigh-Plesset equation which governs the dynamics of a hemispherical bubble [8,9]:

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{1}{\rho}\left(p_{\mathrm{i}} - p_{\infty}(t) - \frac{2\gamma}{R} - \frac{4\mu}{R}\dot{R} + \frac{R}{c}\frac{\mathrm{d}p_{\mathrm{i}}}{\mathrm{d}t} \right) \quad (2)$$

where R is the radius of the hemispherical cap of gas, ρ is the liquid density, γ the surface tension of the liquid, μ the liquid dynamic viscosity, p_{i} the pressure in the bubble and $p_{\infty}(t)$ the liquid pressure taken far from the bubble. This equation cannot be applied as such to gas pockets contained in groove-shaped cavities.

EFFECT OF ULTRASOUND OVER A LONG TIME SCALE:

The purpose of this first study is to examine the effect of ultrasound on gas pockets over a long time scale: about thirty minutes. The aim is to check whether, under these experimental conditions, the ultrasonic waves can degas the interface or, on the contrary, cause the gas pockets to grow by rectified diffusion. The phenomenon of rectified diffusion may induce stabilisation of the bubbles against dissolution or expansion of the bubbles or gas pockets. This process takes place progressively, over several acoustic wave cycles, by preponderant gas diffusion at the liquid/gas interface in the liquid direction towards gas [6]. It is expected the effect of rectified diffusion to be negligible since the used frequency is probably too high to produce significant diffusion of gas in the gas pockets [10]. Due to this phenomenon, it seems very unlikely that coalescence can occur. Whatever the case, if its effect is non-negligible, a decrease in the amplitude of the transmitted ultrasonic signal over time (as the surface fraction of gas at the interface increases) should be observed. The surface fraction of gas at the interface is measured indirectly from the transmitted ultrasonic signal. For example, for a substrate whose surface fraction of purely solid/liquid interface τ is low, the amplitude of the transmitted signal should normally be very low.

Comparative tests were conducted. They consisted in measuring the peak-to-peak amplitude of the transmitted ultrasonic signal over time, for cases with and without ultrasonic waves. In the first series of tests: without ultrasound, a train of ultrasonic waves is emitted every five or ten minutes in order to monitor the changes in the signal over time without having too great an influence on the evolution of the gas pockets. In the second series of tests: with ultrasound, a wave train is emitted every one hundred milliseconds. For all substrates, a progressive increase in the amplitude of the received signals over a long time scale is observed: of the order of several minutes. In addition, the rate of increase in the amplitude in the case where the substrate is subjected to ultrasound is very close to that of the case "without ultrasound" (Fig.4). However, the rate of increase is slightly higher for the case "with ultrasound".

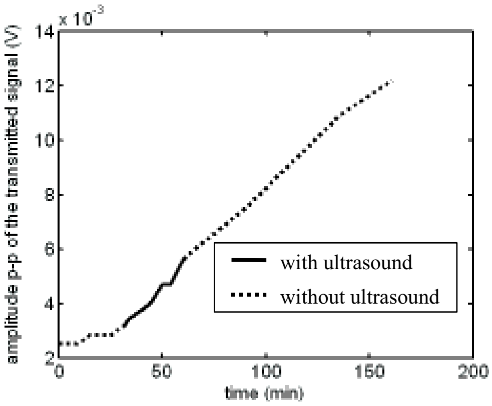

Fig. 4: Graph of the peak-to-peak amplitude of the transmitted ultrasonic signal for substrate S6 against time.

The ultrasound measurements therefore demonstrate a change over time in the amplitude of the transmitted ultrasonic signal opposite to that which the effects of rectified diffusion would produce if its influence was significant. In addition, the images recorded at the same time as the ultrasonic signals on various time scales reveal no perceptible variation in the shape or dimension of the gas pockets. The experimental results therefore confirm the negligible effect of rectified diffusion on the gas pockets.

EFFECT OF THE ULTRASONIC WAVE OVER A SHORT TIME SCALE:

In this second study, the effect of ultrasound on gas pockets over a short time scale, from 1 to 10 µs, is examined. The aim is to check whether, under these experimental conditions, the ultrasonic wave expand the gas pockets sufficiently to induce coalescence. If this phenomenon occurs, by accurately adjusting the experimental synchronisation equipment, it is possible to monitor the expansion of the gas pockets leading to coalescence. If coalescence does not occur, the setup can detect any expansion of the gas pockets, and even an oscillation of the liquid/gas interfaces if it is significant.

The experimental device makes it possible to observe the movement of the liquid/gas interfaces of the gas pockets by temporal scanning of the photographs with respect to the ultrasonic wave. To monitor fairly accurately the changing shape of the interfaces over an ultrasonic wave compression/expansion cycle, a time shift step of 200 ns is chosen. Obviously, without ultrafast camera, this scanning is not carried out on one but several wave trains, triggered manually and therefore with no periodicity. This study is based on the hypothesis that the phenomenon is perfectly reproducible from one wave train to another. This hypothesis was verified by Bremond et al. [7].

The images observed are identical to those obtained without the ultrasonic waves. The image sequences recorded show that the shape of the gas pocket interface is unchanged by the ultrasonic wave. Neither any significant oscillation of the liquid/gas interfaces during the passage of the ultrasonic waves is observed. Under these experimental conditions therefore, the ultrasonic wave do not cause the gas pockets to expand. Coalescence by expansion of the gas pockets according to the gas law (i.e. without gas diffusion) does not occur.

If no movement of the gas pocket interface is observed, this is related to the acoustic pressures involved, estimated to be several thousand Pa, which are therefore relatively low. The gas pocket interfaces must only oscillate very slightly during the passage of the wave. In the experiment conducted by Bremond et al. [7], the depressions generated were about 1000 times greater than those imposed in the present experimental setup.

OBSERVATION OF ANOTHER PHENOMENON:

In a sequence of images of the substrate S6 not subjected to ultrasound (Fig.5), with a time interval of one minute, a groove filling with water is observed. The groove is progressively filled with water, towards the bottom.

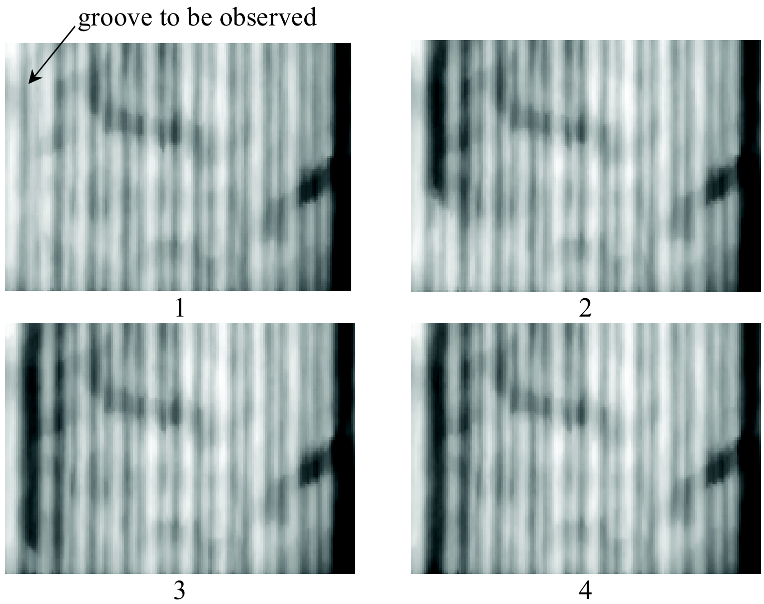

Fig. 5: Sequence of images numbered 1 to 4 of a portion of the surface of substrate S6 (ten vertical grooves) showing a groove filling with water.

The following interpretation is given to explain this observation. It is likely that there are small defects on the surface of the substrates, such as local sensitive variations in the roughness or heterogeneities in the surface chemical treatment, i.e. small non-hydrophobic areas. The presence of a defect on one of the two edges at the top of a groove could cause a very local change in the contact angle and therefore lead to water penetration at this point in the groove. With time, therefore, under the effect of the hydrostatic pressure, water could invade the entire groove. This filling phenomenon can be avoided, for example, by using substrates with cylindrical holes (of diameter as small as possible) rather than grooves in order to minimise the perimeter of the liquid/gas interface. The shorter the triple line (intersection of the three phases: solid, liquid and gas), the lower the risk of finding a defect on this length. Another phenomenon which could explain the grooves filling with water is diffusion of gas at the liquid/gas interface of the pocket towards the liquid under the effect of the liquid pressure at the substrate [10]. In this case, filling would occur homogeneously along the groove.

CONCLUSION:

An experiment has been carried out in order to observe the ultrasound-induced behaviour of gas pockets. The results demonstrated that these experimental conditions are not favourable to the occurrence of coalescence or even to a significant growth of the gas pockets. Over a very short time scale, expansion of the gas pockets according to the gas law is negligible due to the relatively low acoustic pressures involved. Over a long time scale, rectified diffusion has a negligible effect since the frequency of the ultrasonic waves is relatively high. In addition, this experiment allows concluding that the effect of ultrasound on progressive degassing of the substrate surface is negligible. The ultrasonic waves are not responsible for the increase in the transmitted ultrasonic signal over time. This phenomenon corresponds to a progressive filling of the grooves with water. This filling with water is attributed to small variations in the highly local equilibrium of the gas pockets which may be unstable depending on the stability of the hydrophobic layer, the local roughness, the gas diffusion, the hydrostatic and acoustic pressure, etc.

Based on these results, which allow dismissing the coalescence hypothesis, the acoustic transmission through a composite interface is planned to be modelled considering the scattering due to the gas pockets.

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