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Ultrasonic coda wave interferometry (CWI) for detecting a change at interface of a solid surface - Applications for monitoring fouling, biofilm growth, cleaning and corrosion

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ABSTRACT

Fouling of heat exchanger and corrosion occurs in many industrial sectors which causes considerable economic losses and increases public health risks. Therefore, it is important to develop devices to monitor the formation/removal of fouling and the corrosion occurrence. Such devices permit to revisit the fouling/cleaning process and the corrosion sequences in order to minimize associated impacts and losses. In this work, a coda wave interferometry (CWI) based-sensor is used to monitor the fouling and corrosion status of surfaces. This paper aimed to give an overview of the experimental results obtained in order to assess the potential of this technique for various types of applications (wax cleaning, biofilm formation detection, fouling/cleaning monitoring for dairy derivatives-unpreceded results and corrosion formation). Overall, our work provided evidence that CWI method is applicable for monitoring fouling factor and corrosion of solid surfaces and could be easily implemented in a non-invasive way and on line.

INTRODUCTION

Fouling and corrosion of heat exchangers occurs in many industrial sectors such as oil, pharmaceutical and food ones. The generation of unwanted deposits in food processing facilities compromises for example the quality and safety of edible productions. Thus, frequent cleaning cycles have to be regularly planned to achieve hygiene standard of the processing lines, to limit the risk of cross-contamination of products and food poisoning. The cleaning of the fouled surface (to avoid microbiological growth or survival) is generally carried out using water and harsh chemical detergents and could be considered as highly critical as regards sustainability. Indeed, everybody is aware now that these resources e.g. potable water are limited and that public claims green chemistry for detergents. On the other hand, the occurring of corrosion also contributes to the crosscontamination of products and can cause serious harm to the consumer. Therefore, frequent maintenance interventions are needed in order to avoid problems related to food product quality and safety and malfunction of equipment. Key points for cleaning improvement or corrosion control are the detection and monitoring in order to be continuously aware of the current state of equipment surfaces. Such detection permits to optimize the cleaning procedure and minimize the use of detergent but also to select a solid surface integrating corrosion resistance and improve social demand as regards circular economy.

Therefore, it is of importance to develop non-destructive devices able to detect a

change at the interface of a solid surface in order to be well-informed about fouling factor or corrosion status of the surface. The objective of an NDT/E (Non-Destructive Testing/Evaluation) method is to detect and characterize the state of structures or materials, without modifying the studied specimen, which means the methods are certainly non-invasive. Thanks to a good penetration of acoustic waves, ultrasonic methods are widely used to characterize liquid or solid materials since the last century. Among all the NDT/E methods, the ultrasonic coda wave seems to be a suitable method for measuring fouling factor or corrosion status of a surface. Briefly, coda wave is the late part in signals after the direct wave, which propagate directly from acoustic source to the receiver. It is a summation of waves along all possible paths with multiple reflections and diffusions. The detection methods, based on coda wave, are more sensitive than the classic acoustic methods. In fact, coda waves have been widely applied in seismic research. However, this potential has not been evaluated for measuring fouling factor or corrosion status of a surface.

The objectives of this this extended abstract is to highlight that the CWI is applicable to monitor fouling factor or to detect corrosion of the surface. We have gathered here published and original results of our group illustrating how this technique has been implemented for various types of applications (monitoring wax and protein deposit cleaning, detecting biofilm formation and detecting corrosion appearance) and showing its ability to detect change at interface of a solid surface.

Fouling and cleaning results presented here have been mainly obtained in the frame of collaborations between permanent scientists of UMET and UPHF started in 2016 and still ongoing. This collaboration included B. Chen PhD thesis but also the further bringing of Master students and in Post-doctoral positions (A. Boutignon, T. Danel, S. Khelissa and M Abdallah) on hygienic issues in food industry.

Corrosion studies are more recent and still on course and have been acquired in the frame of Interreg SOCORRO2 project (Seeking out corrosion - before it is too late). This work was supported by the European project Interreg 2 Seas, Antwerp Maritime Academy, SOCORRO.

SOCORRO2 still involves many permanent scientists of UMET and UPHF with the help of Post-doctorant positions (C. Nicard, M. Farin, S. Khelissa and M. Abdallah). This European project aims to provide industries with an independent tool based on machine-learning to assess corrosion risks of their installations in contact with sea water. The objective is to rise their awareness and provide them with the appropriate tools in order to take preventive actions against corrosion damage that often results in economic losses.

MATERIALS AND METHODS

More detailed information about materials and methods can be found in original publications [1-6] corresponding to the different presented-applications. For concision and clarity, only essential informations are given here.

Principle of ultrasound coda wave interferometry

In classical ultrasound techniques, echographic measurements use only direct ultrasound waves to detect and characterize changes in medium properties. These waves are the ones that propagate directly from the source to the sensor via a unique reflection at a given interface, and they consequently appear in the first part of the signal recording (basic illustration for detection of soils is shown in Fig. 1-left. In applications, where changes at a solid substrate interface occur, these direct echos will be only very slightly affected, and consequently very low property contrast could not be detected and will make the detection of a change at interface hazardous. On the contrary, multiply-

reflected and multiply-scattered wavepackets will cumulate the effects introduced by the substrate changes over several wave paths (Fig. 1-right). These so-called ‘‘coda waves’’ will naturally appear later signal plot. Briefly, the medium is crossed one time by direct waves and several times by coda waves. This will result in a much higher sensitivity of the coda waves to small property changes (such as caused here by the biofilm formation, Protein deposition, wax detachment) than the direct wavepackets. Similar phenomena are also detected whether substrate is damaged by corrosion occurrence.

Fig. 1. Schematic illustration of ultrasound wave propagation in the medium: direct waves (left) correspond to first reflected and backward propagated echoes, whereas coda waves (right) are multiply reflected and scattered inside the medium when a change at solid interface of substrate occurs.

Since coda corresponds to a superposition of waves propagating along various paths, it is difficult to analyse it using classical acoustic parameters such as time-of-flight, reflection and transmission coefficients. Instead, useful information will be extracted by quantifying coda signal changes or dissimilarities between two states of the medium.

To achieve this goal, the decorrelation coefficient is generally used as an indicator of the dissimilarity between two recorded coda signals s_1 and s_2, respectively. One of the two signals is the reference one. This dissimilarity is computed in a given time-window $[t_0, t_1]$ according to the following equation [7]:

$$D_{1,2} = 1 - \frac{\int_{t_0}^{t_1} s_1 s_2 dt}{\sqrt{\int_{t_0}^{t_1} s_1^2 dt \int_{t_0}^{t_1} s_2^2 dt}} \quad (1)$$

The second term in Eq. (1) corresponds to the normalized cross-correlation of s_1 and s_2 at zero lapse-time. Mathematically, the value of $D_{1,2}$ is between 0 and 2. Identical s_1 and s_2 signals lead to zero decorrelation coefficient and nonzero values are an indicator of waveform shapes and phases differences between signals. Hence, the value of $D_{1,2}$ will be directly related to the degrees of state changes in the medium. Therefore, in our application, this evolution during the monitoring procedure is expected to be linked to soil attachment/detachment on the substrate or damage of the substrate by corrosion.

Sensors and acquisition chain required for coda wave interferometry

The transducers are needed to emit and receive the ultrasonic signal used in these experiments. They are low-cost piezoelectric patches (Fig. 2), consisting of two separate electroded parts for emission and reception. The transducers are glued on the lower side of the substrate (solid surface) for which a change at upper side interface need to be monitored. The acoustic sensor on the fouled substrate is connected to an acquisition system employed to generate, acquire and process the signal. This acquisition system consists of 3 parts (Acoustic Generator; Acquisition Board; Amplifier). Excitation strategy (frequency band of the emission signal) requires to be fixed according to the characteristic of the solid substrate for which a change-detection needs to be monitored.

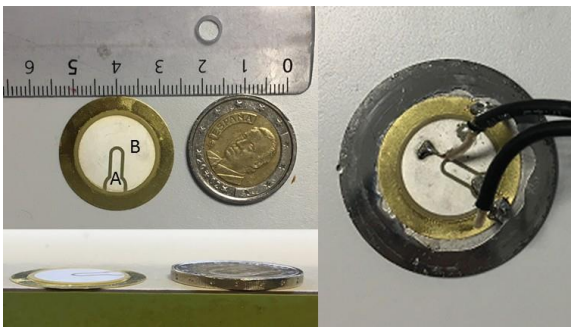

Fig. 2. Piezoelectric transducer consisting of two parts: Receiver (A); Source (B).

A know-how is also necessary to select the best interval of time (window) for analyzing the coda wave signal as the sensibility is poor in the earliest (direct wave) and too affected by experimental noise in the latest instant.

Cleaning of wax deposited on a stainless-steel substrate

A piece of wax is deposited on the inner surface of a rectangular channel in a stainless steel to mimic the presence of a fouling layer on a substrate. Two ultrasonic sensors are installed at the outer face of the substrate; one in the vicinity where the wax has been deposited while the other one is located at the outer face of the clean substrate as a witness (control). Hot water (70°C) is used to flush out the wax in the channel as a cleaning procedure. This cleaning process of wax is monitored by CWI method. The difference between the monitoring results for the 2 sensors (with and without wax) is then discussed.

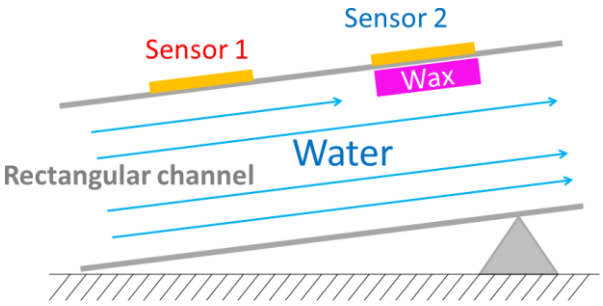

Fig. 3. Diagram detailing the inside and the outside of the duct (laboratory scale). The hot waterflows through the duct from left to right.

Detection of the biofilm formation on a stainless-steel substrate

Bacteria are used to generate a biofilm layer on a substrate within 2 days. For performing that, a certain quantity of bacteria (*Staphylococcus aureus*) and nutrient solution is inoculated into a previously sterilized container. Then, the containers are incubated in a thermostatic stove at 30°C. Another identical sterilized container was only filled with nutrient solution and used as a witness (control). The evolution of the monitoring obtained by CWI for the 2 containers (with and without bacteria-Fig. 4) is then discussed.

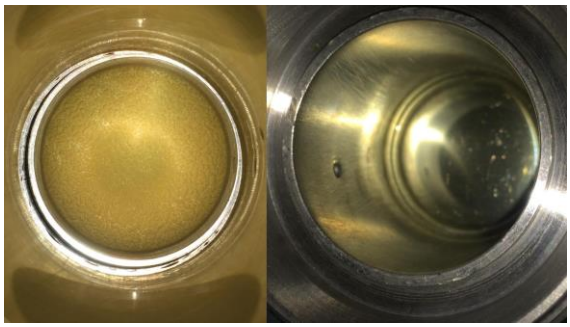

Fig. 4. Photo of biofilm formed on the substrate of Container 1 (Left); Photo of clean substrate of Container 2 (Right)

Cleaning of denaturated protein on a stainless-steel substrate

Firstly, a layer of whey protein deposit is generated on a stainless-steel coupon by a homemade device. For that, the coupon is immerged in a sealed tube containing 5% w/w of Whey Protein solution and 500ppm of $CaCl_2$. The sealed tube can rotate and is placed at 80°C in a thermostat cabinet. Then the coupon for which an ultrasonic sensor has been glued is placed is in a sample tube (Fig.5). A 3 steps cleaning sequence is applied on the sample tube at a given temperature (50°C) and a given flow rate (100 l/h). Water is used as cleaning agent on step 1 (pre-rinsing) and step 3 (post-rinsing). In step 2, the cleaning process is performed by circulation of sodium hydroxide.

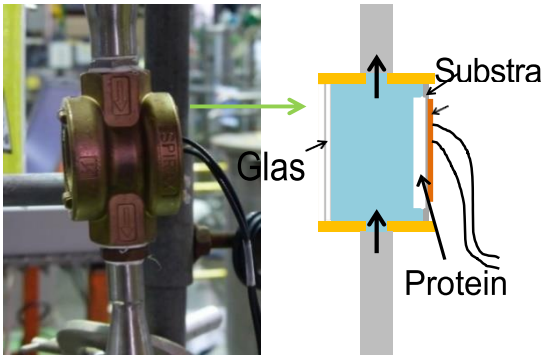

Fig. 5. Sample tube placed in a Cleaning In Place device containing the substrate fouled with protein deposit and on which an ultrasonic sensor is embedded.

An optical observing is carried on during the whole measurements by a camera placed in front of the glass. The video is sampled in images and grey level is analyzed to get information of the fouling status. The final state is taken as the reference state for evaluating the decorrelation coefficient evolution.

Corrosion detection on S355 steel substrate

Corrosion experiments have been performed by immerging S355 steel in Fresh-water and Salt-water (**NaCl 3 wt.%) (Fig. 6)**. The electrolyte is temperature controlled. Iron release kinetic and pictures of corroded surface have also been followed.

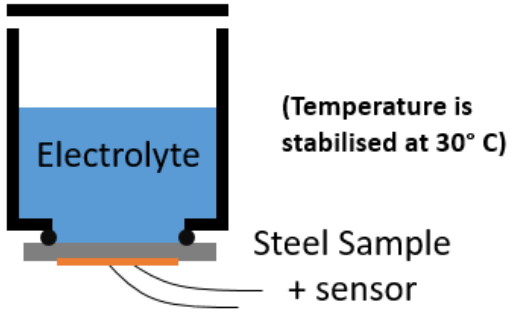

Fig. 6. Sample tube placed in a Cleaning In Place device containing the substrate fouled with protein deposit and on which an ultrasonic sensor is embedded.

RESULTS

Cleaning of wax deposited on a stainless-steel substrate

For the sensor in the vicinity of wax (blue line), Fig. 7 firstly shows a sharp increase of decorrelation coefficient with cleaning time which was followed by an asymptotic value after 1h. However, few evolutions of decorrelation coefficient are noted for the control sensor (orange line). The evolution of decorrelation coefficient is correlated with wax detachment which was confirmed by the images acquired using the same conditions fouling/cleaning with a similar device covered with a transparent duct.

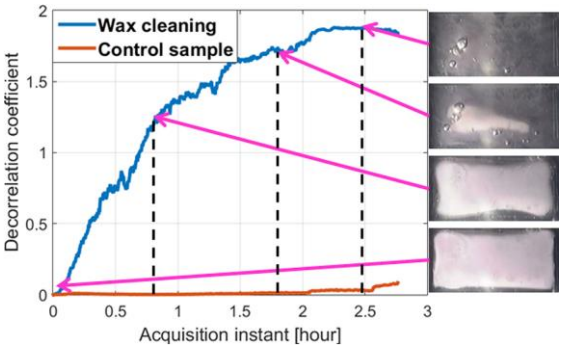

Fig. 7. Decorrelation coefficient curves: control sample (red) and wax cleaning process (blue) for the window 16–26 µs. Photo of wax at different moments of cleaning protocol (right,from bottom to top): reference state (soiled by wax) , state after 0.8 h of cleaning, state at 1.8 h of cleaning, state after 2.5 h (clean).

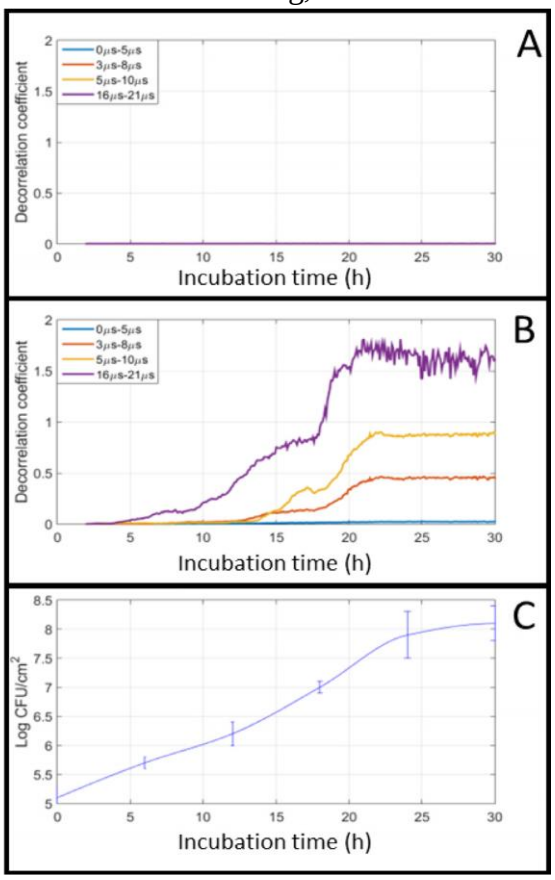

Fig. 8. Decorrelation coefficient evolution as a function of incubation time for: A presents the control sample, B presents the biofilm formation sample, with different time windows: 0 µs − 5 µs, 3 µs − 8 µs, 5 µs − 10 µs, 16 µs − 21 µs (frequency band 9 MHz–11 MHz); C presents the kinetic of *S.*

aureus biofilm formation on stainless steel substrate.

Formation of a biofilm on a stainless-steel substrate

In the negative control container (Fig. 8A), the decorrelation coefficients are close to 0 for any incubation time and selected time window. On the contrary, the results in Fig. 8B show that the decorrelation coefficients of biofilm monitoring sample increased significantly as the incubation time increased. It also shows that the sensitivity of the state change detection depends on the time windows. Fig. 8C shows that the biomass evolution could be compared with the decorrelation coefficient. Indeed, the data of the bacterial cell enumeration seems in agreement with the decorrelation coefficient evolution.

Cleaning of denaturated protein on a stainless-steel substrate

Fig. 9 (Blue curve) shows the evolution of decorrelation coefficient during the cleaning cycle. The decorrelation coefficient stays stable for the first 10 minutes, which is the pre-cleaning rinsing step. For the fouling cleaning step (10-35 minutes), the decorrelation coefficient decreases to 0 within 4 minutes. Then it stays at zero till the end of step 3. These evolutions agree with the diagnosis obtained by images analysis. The fouling state does not change at the first step which is logic since the water alone is not supposed to be efficient enough to get rid of the protein deposit. During the cleaning step, the video shows that the fouling layer is eliminated rapidly and the substrate become clean after about 3.5 minutes. As the fouling layer has already been totally eliminated at that moment, it is logical that there is no more evolution during the step 3 (Post rinsing step).

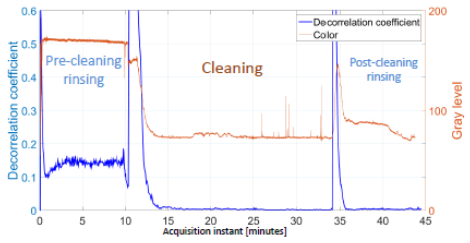

Fig. 9. The decorrelation coefficient (Blue) and Gray level evolutions during cleaning of proteinaceous deposits (Red).

Corrosion formation on S355 substrate:

Fig. 10 shows that no evolution of decorrelation coefficient is observed when sample is in air (no or negligible corrosion). However, stronger decorrelation coefficient is noticeable when sample is in contact with salted water rather than unsalted water.

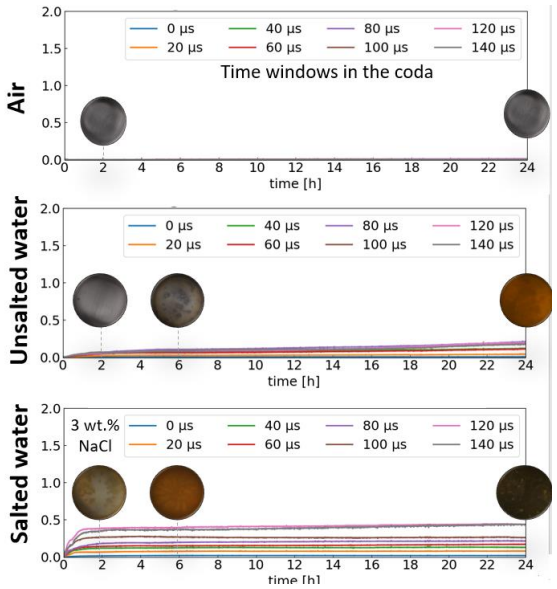

Fig. 10. The decorrelation coefficient evolutions for a steel substrate placed in various medium (air, immerged in unsalted water and in salted water)

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Conclusion

Overall, this work has provided evidence that the proposed-CWI method is a good

option to detect a change at interface of a solid surface in order to get information about fouling factor or corrosion status of a surface. It is worth mentioning that a lot of research are still on going. For example, now a set up to perform CWI measurements of corrosion electrochemically controlled is possible and under investigation. It should be noted that the presented results are based to the evolution of decorrelation coefficient. However, our efforts are now focused on other indicators such as the dilatation coefficient which seems to be relevant to analyse CWI results [6]. Indeed, CWI technique is sensitive to temperature changes since this parameter affects the ultrasound coda properties. Consequently, problems in applications can come up and limit the extension of CWI methods in cases where significant temperature changes cannot be controlled as for example detecting corrosion in sea water and detecting fouling status in real CIP devices as cleaning solutions (pre-rinsing water, caustic soda, acid, rinsing water) doesn't operate at similar temperature as heat treated product.

In this case, we have noticed that dilatation coefficient can constitute a possible way to consider the impact of temperature changes and to improve the accuracy of detection. This work is currently still under progress [6]. Furthermore, our research is now devoted to the improvement of this method to be able to detect corrosion induced by biofilms using the marine model.

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