Deliverable 3.3

EC's Framework Programme for Research and Innovation Horizon 2020 (2014-2020)
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Cross-sectorial real-time sensing, advanced control and optimisation of batch processes saving energy and raw materials (RECOBA)

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Acoustic sensor for particle morphology monitoring

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Lead contractor for this deliverable:	UCAM

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Dissimination level

PU	public	☒
PP	restricted to other programme participants (incl. the Commission Services)	☐
RE	restricted to a group specified by the consortium (incl. the Commission Services)	☐
CO	Confidential, only for members of the consortium (incl. the Commission Services)	☐

CONTENT

1 Introduction

The aim of deliverable 3.3 is to describe the progress made and the results achieved on particle sensing using thin film bulk acoustic wave resonator (FBAR) technology. The work performed has been mainly focused on two different areas:

1. Fabrication of the piezoelectric FBAR transducer
2. Sensing experiments with solutions containing polymer particles

While the work on the fabrication of the piezoelectric FBAR transducer was performed in the laboratories of the University of Cambridge, the sensing experiments took place in the Universidad Politécnica de Madrid, in the facilities of the Group of Microsystems and Electronic Materials (GMME), during the first two weeks of December, enabled by a COST Action STSM. As it will be described in the corresponding section, they have set up a custom-made experimental arrangement that provides a reliable way to perform sensing experiments and handling liquid samples using FBARs.

2 Fabrication of the piezoelectric FBAR transducer

2.1 Deposition of ZnO layers

The use of FBAR as sensors is based on the change in their resonant frequency that occurs when a mass is attached to their surface, also known as gravimetric sensing. The resonance is achieved through the conversion of electrical energy applied to the device into mechanical energy (and vice versa) thanks to a thin piezoelectric layer, usually zinc oxide (ZnO) or aluminium nitride (AlN), that is sputter-deposited between the metallic electrodes. There are two main parameters that will indicate the goodness of a FBAR device: the electromechanical coupling factor (k^2) and the quality factor at the resonant frequency (Q). The first one indicates the efficiency of the conversion between the electrical energy applied to the device and the acoustic energy generated by the piezoelectric layer. It depends mainly on the quality of this piezoelectric layer. The second parameter, the quality factor, is related to the energy losses suffered by the resonator and depends on many factors, such as the contacts, the materials used in the device, its geometry, etc. The Q is the critical parameter in resonators intended for sensing applications, because it is related to the sharpness of the resonant peak. If it is not sharp enough it is extremely difficult to determine the value of the resonant frequency accurately. In that case it is not possible to track it; therefore identifying the shifts related to the mass being bond to the surface of the transducer is not feasible.

FBARs display two different modes of operation depending on the direction of the vibration of the particles with respect to the direction of propagation of the wave. If the vibration is perpendicular to the propagation, we will refer to the mode as the ‘shear’ mode. If it is parallel, the mode will be the ‘longitudinal’. The shear mode has a particularity that makes it especially suitable for in liquid sensing: it does not couple into the liquids, which means that it suffers a much lower attenuation (maintains a higher Q in liquid compared to the longitudinal mode). Figure 1 displays the frequency response of a resonator showing both modes and operating in air and in liquid.

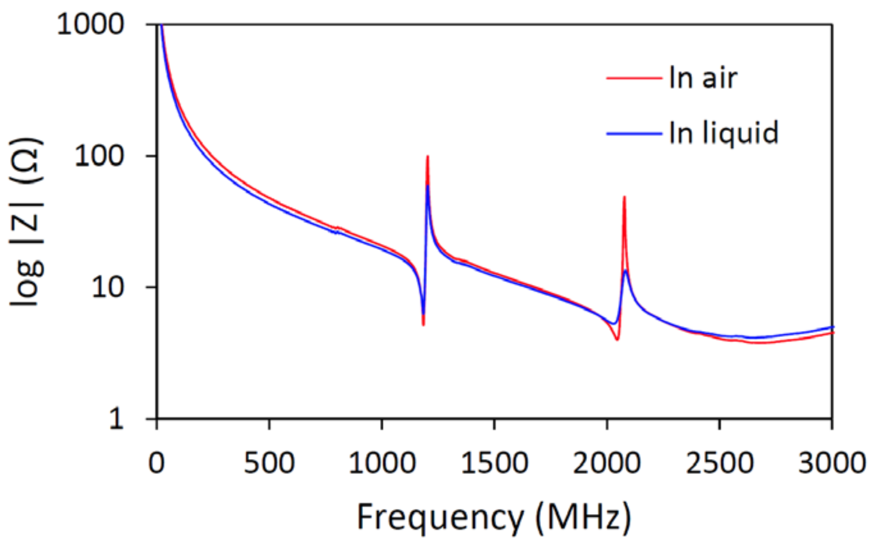

Figure 1 – FBAR displaying the shear (at ~1200 MHz) and the longitudinal mode (at ~2050 MHz) operating in air and in liquid.

While the shear mode is capable of maintaining a $Q \approx 150$ in liquid, the Q of the longitudinal mode reduces to values below 10, which is insufficient for sensing purposes.

The requirements set by the RECOBA project involve the detection of the particles in fluids, which essentially means that the FBARs must operate in the shear mode. However, the excitation of this mode is not straightforward, and is dependent on the microstructure of the piezoelectric materials. We have optimized a process for producing piezoelectric ZnO films with a microstructure to yield FBAR devices with the response shown in figure 2.

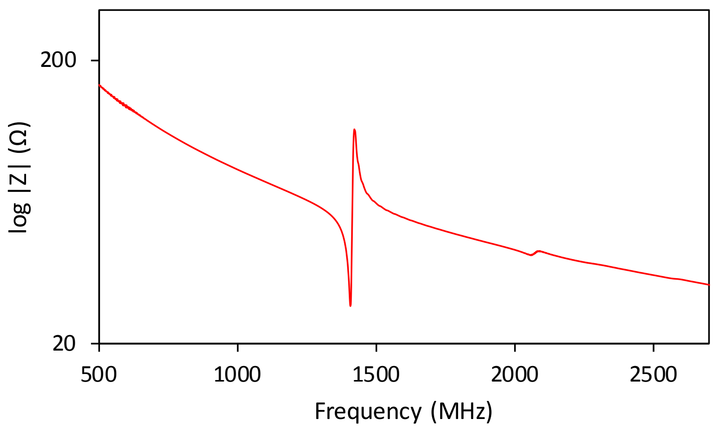

Figure 2 – FBAR operating in the shear mode and fabricated using ZnO thin films with an optimised microstructure.

The devices fabricated display a k^2_{shear}≈2.8% and a Q≈120, which ensures that the resonant peak is sharp enough to determine the resonant frequency accurately.

2.2 Design of the test transducers

Another challenge derived from the requirement of the operation in liquids is the actual handling of the samples and the compatibility with the devices. The design of the resonators should enable the fitting of a fluidic system that keeps the sample away from the measurement probes. This means that the contacts to the active area must be extended. Given the high frequency of operation of the FBARs, above 1 GHz, such extensions should be carefully designed to control the parasitic elements they add to the equivalent circuit of the device and that can cause serious damage to the resonance [1]. This has been achieved using a specific FBAR device structure and microfluidic system which is ready to be attached to a measurement setup to perform particulate detection.

3 Sensing experiments with solutions containing polymer particles

As it has been stated in the introduction, the sensing experiments were performed in the laboratories of the GMME in Madrid. The sensor is attached to the closed fluidic circuit that flows the samples using a fluidic pump. Figure 3 shows a picture of the setup, with the different parts highlighted.

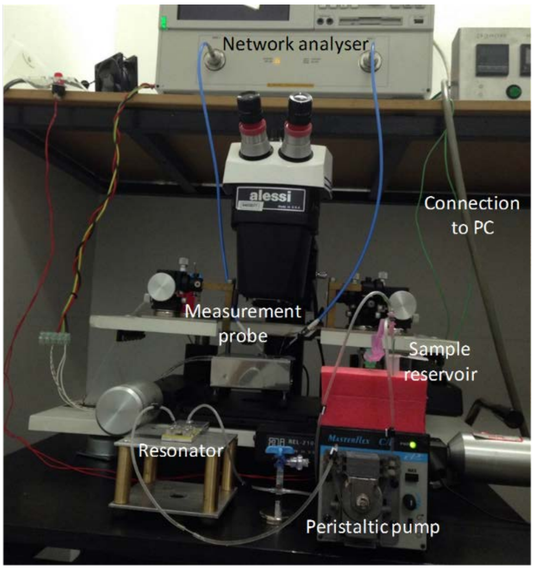

Figure 3 – Picture of the experimental setup with closed fluidic circuit.

The experimental setup enables recirculation of the samples if necessary. The tracking of the resonant frequency in time is performed using ad-hoc LabVIEW software that determines accurately the resonant frequency from the frequency response acquired with the network analyser and represents its evolution in time.

The polymer samples used for the experiments were provided by Polymat. They are structured particles with a core-shell structure. The test solutions where prepared from the samples diluted in deionised (DI) water. Prior to the feeding of the prepared sample DI water was introduced in the circuit while tracking the resonant frequency to stablish a baseline. Figure 4 shows the result of one of the described experiments.

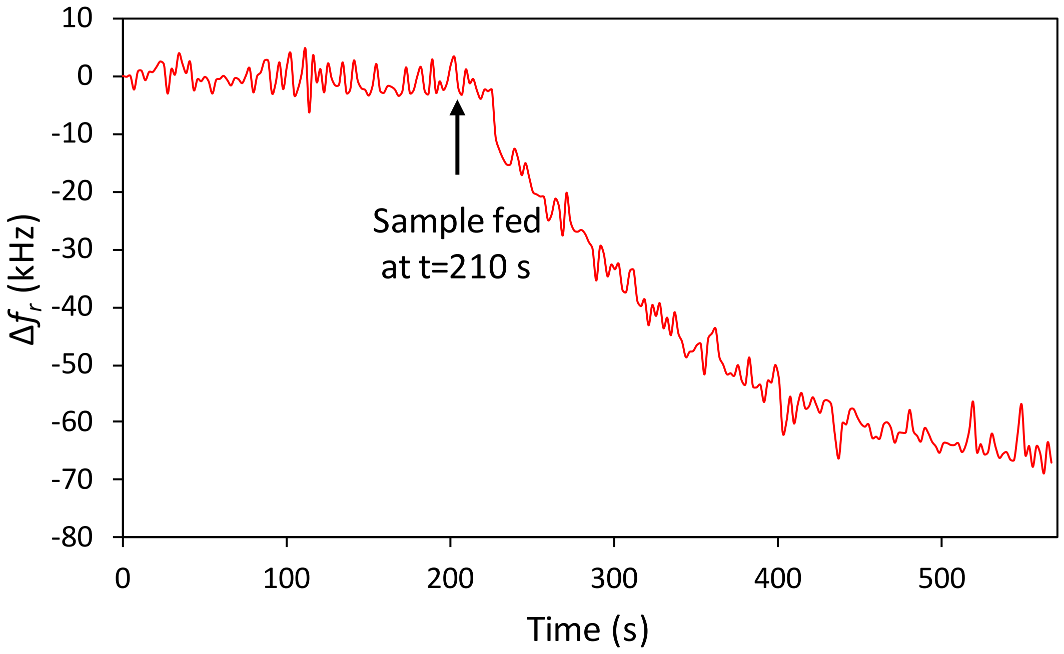

Figure 4 – Evolution of the resonant frequency of the transducer during the experiment.

When the sample contacts the active area of the device a progressive shift takes place until saturation. From previous experiences with this kind of devices, such a shift cannot be attributed to a change of the viscosity of the liquid, which suggests that the particulates must be binding to the active area. Rinsing with DI water with the same rate does not detach the particles.

Even though these experiments only constitute a proof of concept, they are extremely promising. The detection of the particles seems to be possible using FBARs in the light of the presented results. New experiments will be performed in the coming months using solutions with different types of particles, with the objective of finding a correlation between the frequency shift and the morphology (at least the size if not the shape) of the particulates.

4 Conclusions

As a summary of this deliverable 3.3 the following conclusions can be drawn:

- A novel method to deposit microstructured ZnO thin films has been developed. Using this method, it is possible to fabricate shear mode FBARs that display a k^2_{shear}≈2.8% and a Q≈120. Test transducers have been designed and fabricated to fit a custom made fluidic system necessary to handle the liquid samples.
- Sensing experiments of polymer particulates have been performed using FBARs with successful results. They constitute a proof of concept and further experiments need to be perform, but they are very promising and indicate that the detection is possible using FBARs.

The next months (M13-M19) will be focused on the development of surface acoustic wave devices (SAW) for the identification of the morphology of the particles. In parallel the work on the FBAR technology will continue to optimise the design of the transducers and test their response to different type of particles.

5 References

[1] M. DeMiguel-Ramos, M. Barba, T. Mirea, J. Olivares, M. Clement, J. Sangrador, and E. Iborra, “Influence of the electrical extensions in AlN-BAW resonators for in-liquid biosensors,” in *Proceedings - European Frequency and Time Forum (EFTF)*, 2014, pp. 301–304.

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Cross-sectorial real-time sensing, advanced control and optimisation of batch processes saving energy and raw materials (RECOBA)

Start of the project:	Jan 1st, 2015
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Acoustic sensor for particle morphology monitoring: second generation

Due date:	June 30, 2016
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Lead contractor for this deliverable:	UCAM

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Dissimination level

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CONTENT

1 Introduction

The aim of deliverable 3.3 (M18) is to describe the progress made and the results achieved on particle sensing using surface acoustic wave (SAW) sensors, which are the second generation of acoustic sensors proposed in RECOBA. SAW sensors base their operation on the shifts in phase and amplitude that a surface wave suffers when it encounters variations in the environment through which it is transmitted. The generation of the acoustic wave is achieved by the combination of interdigitated transducers (IDTs) and a piezoelectric material. The geometry of the IDTs will determine the electrical response of the SAW device, along with the material properties of the piezoelectric layer [1]. We have chosen a delay line configuration for the SAW sensors, which consists of two sets of IDTs (input and output) with a space between them deposited on top of a ZnO piezoelectric layer. That space is precisely the sensing area where the sample to analyse is placed. The variations of the environment between the input and output will affect the response of the device. Figure 1 displays a schematic of a delay line sensor with the different design parameters that need to be selected.

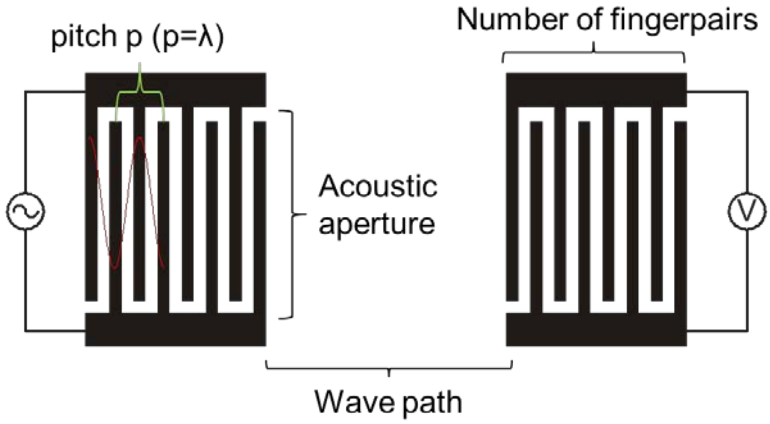

Figure 1 –Sketch of a delay line with the design parameters.

Between M12 and M18 the work performed on SAW sensors has been mainly focused on two different areas:

1. Finite element modelling (FEM) simulations of SAW delay line sensors with textured surfaces for particle detection
2. Design, fabrication and testing of SAW delay line sensors for particle detection

A summary of the results will be presented in the following sections.

2 SAW sensors: FEM simulation

FEM simulations performed using COMSOL Multiphysics ® have been used to analyse the behavior of the acoustic waves in SAW devices and to explore the effects that surface texturing have in their response. The simulated resonators consist of a 5 µm thick ZnO layer on a 40 µm thick Si substrate and Al interdigitated electrodes (IDTs) which are 6 µm wide with a pitch of 24 µm. The model also considers water as the media in contact with the surface of the device. Figure 2 displays the displacement field of one of the simulations.

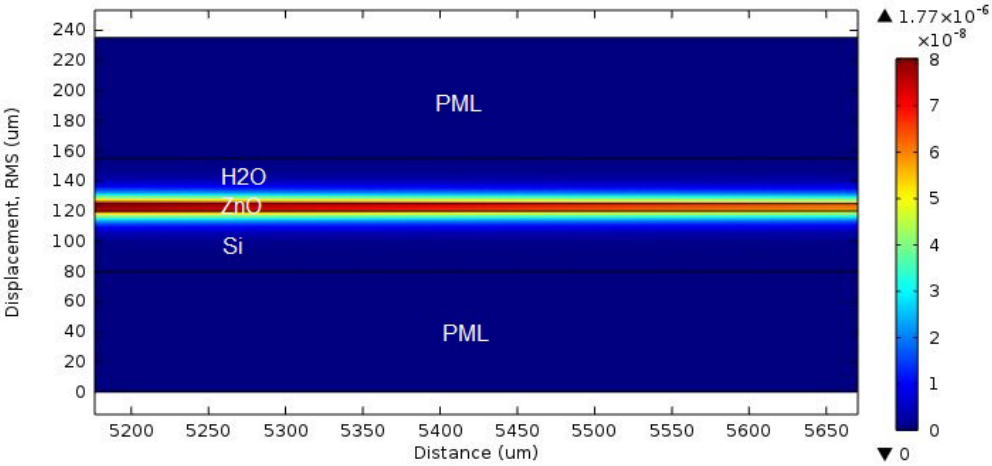

Figure 2 – RMS displacement map in a plain SAW device. The acoustic energy is concentrated in the piezoelectric layer, with exponentially decaying tails in the water and the substrate. The coloured displacement scale is in metres.

Different models have been simulated to study the performance and response of SAW devices with different structures that are micro textured over their surface area. Both 2D and 3D models have been developed, using different structures to texture the surface, namely fins, wells and channels as illustrated in figure 3.

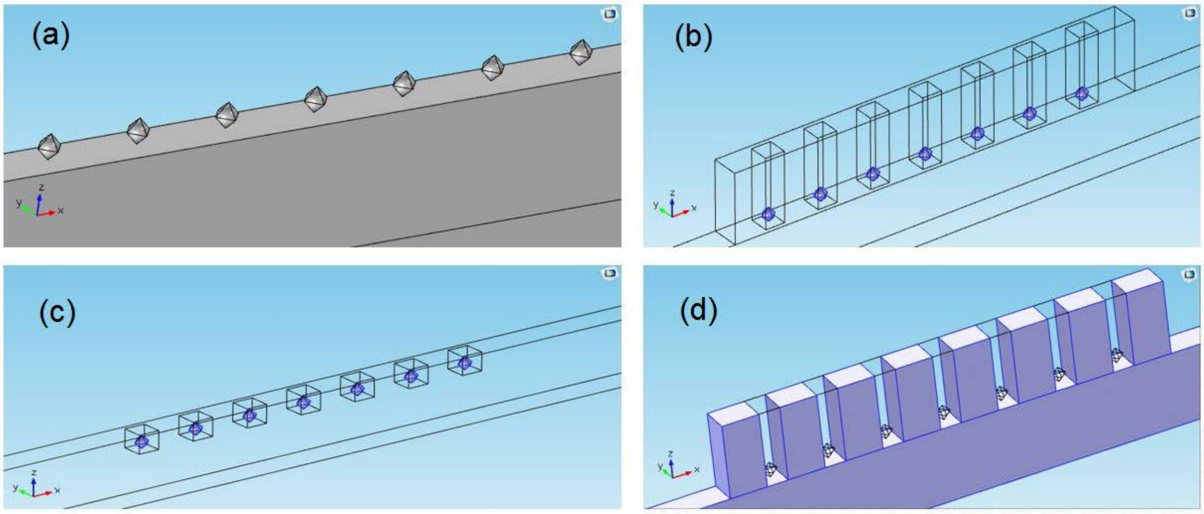

Figure 3 – Surface texturing options simulated with 3D FEM numerical methods: flat surface (a), external wells (b), internal wells (c), and channels (d). In all cases, surface particulates are included in the simulations.

The phase shift in the response of the resonators with different structures depending on the size of the particles can be observed in figure 4. Channels (figure 1-d) display the highest increase of average sensitivity compared to the flat devices (figure 1-a), by approximately 50%. The average sensitivity of this architecture is approximately 0.1° /10 nm radius over the test range.

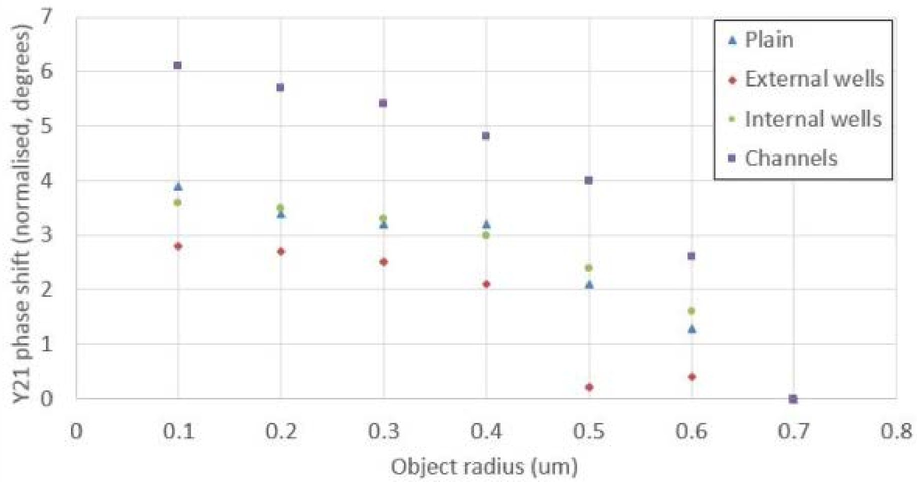

Figure 4 – Summary of results from 3D simulations for various surface texturing options. Phase shift was normalised to 0° for 700 nm object radius to facilitate sensitivity comparison.

The influence of the shape of the particles on the response of the sensors has also been studied and the results of the simulation indicate that this shape also affects the phase shift, which suggests that they can be used to obtain morphological information.

3 SAW sensors: experimental

In addition to the simulation, different versions of SAW delay line sensors have been designed, fabricated and tested. The parameters that have been varied in the different designs are:

- Pitch: 24 µm, 48 µm.
- Number of fingerpairs: 16, 32, 64
- Acoustic aperture: 0.8 mm, 1.2 mm, 1.5 mm
- Wave path: 1.2 mm, 2.4 mm, 4.8 mm, 7.2 mm

The fabricated devices consist of a 500 µm silicon wafer with a thermally grown 2 µm SiO_2 layer. On top of it, a high quality piezoelectric ZnO layer 5 µm thick is sputtered. The IDTs are made of sputtered Al which is 150 nm thick. Figure 5 displays a chip with four different SAW sensors and detail of the IDTs.

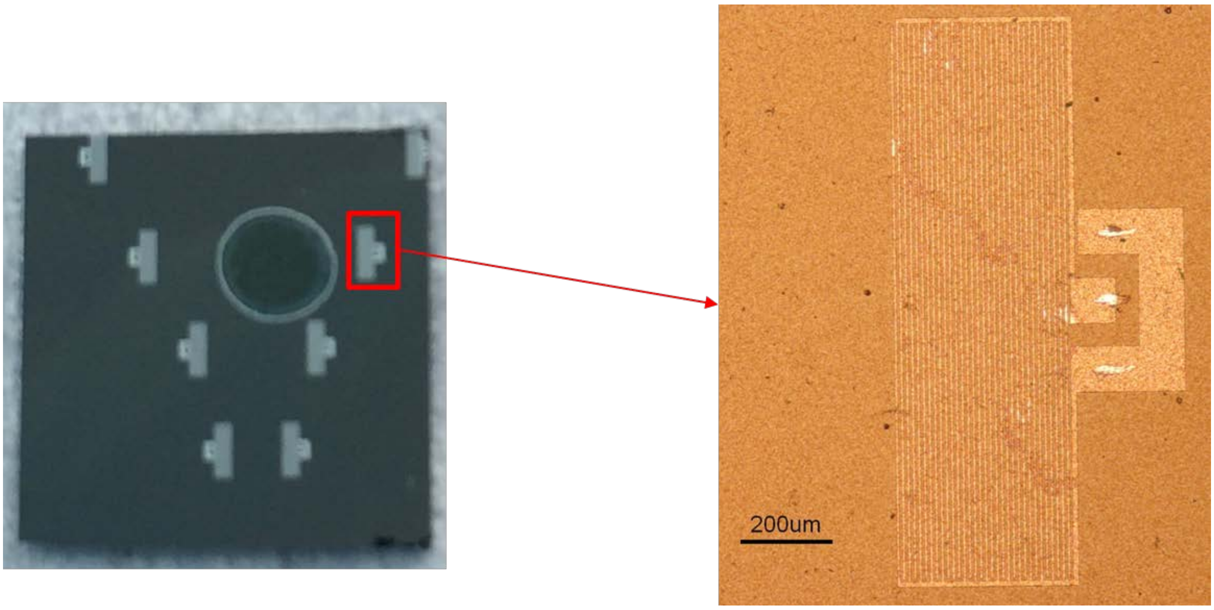

Figure 5 – Chip with four SAW delay line sensors and detail of the IDTs (pitch = 24 µm, acoustic aperture 1.2 mm, number of finger pairs = 16) . The length of the sensing area varies in the different sensors (1.2 mm, 2.4 mm, 4.8 mm, 7.2 mm). A dried drop of emulsion used for the tests can be seen in the delay line.

These devices have been used to perform sensing experiments. Two different types of particles, core-shell (SP1) and raspberry-type (UC4) have been provided in suspension by Polymat and the Department of Chemical Engineering of UCAM. A dilution in DI water with concentration 1:100 has been prepared with each of the emulsions and drops of the solution have been placed in the delay line. The response of the devices displays differences in amplitude and phase of the transmitted signal for the different solutions, as it can be seen in figure 6.

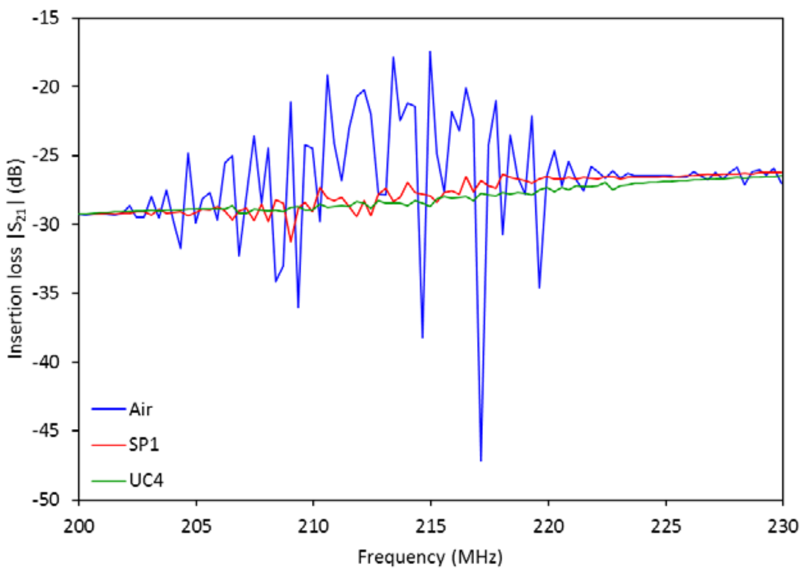

Figure 6 – Electrical response of SAW sensors exposed to air and to two samples containing different types of particles (SP1 and UC4). Despite the high attenuation, different shifts of the resonant frequency for each sample can be observed (SP1 ≈ 5 MHz, UC4 ≈ 2 MHz).

The attenuation of the signal is higher than expected, especially for the second type of solution (UC4). Nevertheless, the differences in the response of the sensors for different solutions suggest that the type of particles may affect the output differently and that SAW devices can be used to obtain some morphological information.

4 Conclusions

As a summary of this deliverable 3.3 (M18) the following conclusions can be drawn:

- The FEM simulations show very interesting and promising results regarding the use of micro textured channels in the sensing area, with a potential increase of the sensitivity by 50%. These results have to be experimentally validated
- The experiments with fabricated sensors indicate that morphological information from the particles may be obtained with this generation of sensors, but more experiments are needed to verify these results despite it being clear that the presence of particles can be detected. In addition to this, some other difficulties are envisaged: the attenuation of the signal is very high when exposed to the samples, which may require the use of other structures (Love mode devices) [2] to obtain sufficiently accurate measurements.

The next months (M18-M30) will be focused on the development of the third acoustic sensor technology, the Angular Acoustic Reflection Spectroscopy (AARS). In parallel the work on the SAW technology will continue, to assess its full potential to fulfill the requirements of the RECOBA project.

5 References

[1] Jr., D. B., Martin, S., Ricco, A., Frye, G., Wohlt-jen, H., White, R., and Zellers, E. Acoustic wave sensors: theory, design, and physico-chemical applications. San Diego: Academic Press, 1997.

[2] K. Kalantar-Zadeh, W. Wlodarski, Y. Y. Chen, B. N. Fry, and K. Galatsis, “Novel Love mode surface acoustic wave based immunosensors,” Sensors Actuators B Chem., vol. 91, no. 1–3, pp. 143–147, Jun. 2003.