Measurement of PM10 and PM2.5 from silicon carbide particles with cascade impactor based on Surface Acoustic Waves sensors

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Abstract— In this study, we used surface acoustic wave (SAW) sensors based on a delay line configuration for measuring particulates matter (PM10 and PM2.5) mass concentration. These sensors exploit the gravimetric sensitivity of Love waves and operate at 125 MHz. It's built on AT-cut quartz and use a silica layer as a guide for Love waves. The classification of particles based on their aerodynamic size was achieved using a custom-designed cascade impactor. The particles collected on the surface of the SAW sensor cause a gravimetric effect that changes the Love wave's phase velocity revealed by a shift of the phase. Hence, monitoring the latter allows the measurement of the mass deposited on the sensor. Our SAW sensors proved its ability to measure PM10 and PM2.5 originated from candle fumes in the 0-500 µg.m⁻³ concentration range. In this paper, we present real time measurements of calibrated Silicon Carbide (SiC) solid.

Keywords—Surface Acoustic Waves, sensors, delay lines, PM10, PM2.5, fines particles, pollution, cascade impactor, air monitoring.

I. INTRODUCTION (HEADING 1)

The first major pollution seems to have been induced by the industrial revolution, allowed by steam engines and coal. In our modern societies, the degradation of our ecosystem continues to increase. The particulate matter PM10 and PM2.5 are very dangerous for human health [1]. Commercialized systems for monitoring air quality are very expensive and cumbersome. A new system based on Love wave SAW sensors combined with cascade impactor has been fabricated in our team. Its ability to separate by size and measure PM10 and PM2.5 in real time has

been proved with particles from candle fumes [2]. As shown in figure 1 and figure 2, the cascade impactor is composed of three stages and work at 3 Lpm flow rate. Coarse particles, exhibiting a diameter higher than 10 μ m, are stopped by the first stage without being measured. The next stage PM10 is dedicated for collecting and measuring particles with 2.5 μ m to 10 μ m diameter. The last stage PM2.5 collects and measures particles with 2.5 μ m to 0.3 μ m diameter. Preliminary experiments using particles from burning candles have been made to show the ability of our system to segregate and measure PM10 and PM2.5. However, it is necessary to check the capabilities of our system to characterize a complex aerosol. In this way, we investigate other type of microparticles such as silicon carbide (SiC).



Fig. 1: Picture of the cascade impactor prototype with integrated SAW sensors



Fig. 2: Schematic of the system based on cascade impactor and SAW sensors as collection plates.

For that purpose, we integrated a calibrated particle generator and an optical granulometer in our experimental setup to generate and characterize complex aerosol.

A. Motivations

Particles with diameter less than 2.5 µm and 10 µm, respectively PM 2.5 and PM10, affect dangerously the human health as they can penetrate deeply in the human respiratory system. The devices used to determine particle concentration are expensive and very cumbersome. The use of micro technology for fine particle detection offers many improvements concerning the size, cost and sensitivity. In particular, Love wave based surface acoustic wave sensors appear as promising candidates for this purpose due to their high sensitivity (250 cm²/g). Despite that, SAW devices are basically not able to select particles according to their diameters. To achieve segregation of particles, a supplementary system is required before measuring. Cascade impactors are widespread for aerosol segregation. However, in order to get proper measurements, the impaction plate is removed and weighted before and after sampling the aerosol, which requires periodic handling. Moreover, a conventional impactor delivers an average measurement through a large sampling time, preventing a real time analysis of a variable aerosol.

B. SAW sensor description

The SAW sensors used are built on an AT-cut quartz. These substrates are characterized by a low frequency-temperature coefficient (FTC) [3] making it appropriate for sensing purpose. They are based on a delay line configuration using IDTs including double fingers 200 nm thick aluminium electrodes. The propagation of Love wave needs a guiding layer. For that purpose, a silica layer was deposited using sputtering technique with shear velocity (*Vs*) lower than the piezoelectric substrates. A schematic of the sensors is shown on figure 3. The perturbations like mass loading involve the condition of propagation. Consequently, the wave phase velocity decreases. Since the wavelength is constant $\lambda = 40 \ \mu m$, the synchronous

frequency will be affected linearly with the variation of the wave phase velocity. This frequency shift is calculated by monitoring the phase signal with a dedicated interrogation electronics [4].



Fig. 3: Design of a SAW delay line based on Love waves $\lambda = 40 \ \mu m \ p = 10 \ \mu m$

C. Custom-designed cascade impactor

The particle separation system used in this work consisted of a custom-designed cascade impactor. With this system, the air sample charged with particles moves across nozzles increasing the flow velocity. When the airflow arrives at the impaction plate, placed below the nozzles, the higher inertial particles diverge from the streamlines and stick on the plate. The stages are typified by the cut-off diameter d_{50} which is equivalent to 50% collection efficiency that rely on the flow rate and the geometry of the system.

The innovative aspect of our impactor is that each impaction plate is equipped with a SAW sensor to measure the mass of particles collected on the different stage of the system. Furthermore, a differential measurement was adopted using a couple of identical devices. One delay line serves to measure particles and the other one is kept clean to work as a reference. This configuration allows to get rid of any contribution of external parameters. As shown in figure 4, previous experiments allowed real-time monitoring of particles from candle fumes and soldering iron.

Particles suspended in the environment have a multitude of origin. It seemed necessary to continue this investigation by testing the system in the presence of carbonaceous particles. These being generated this time by a calibrated system and measured in parallel with a performant optical system. For this, a specific bench of measure has been developed in the laboratory. This allowed showing the correlation between the response of the sensor in phase and the actual particle's concentration in the test chamber. In the next section, we describe the new microparticles test bench developed specifically in the laboratory.



Fig. 4: SAW sensor phase response throughout consecutive exposures to PM2.5 and PM10: the PM10 stage (in green) and the PM2.5 stage (in blue).

D. Microparticles Generation system

To validate the preliminary tests, we have developed a test bench, which will allow us to study other type of particles and control the conditions of tests. The particles size distribution and concentration should be maintained constant during the test time. The candle fumes did not respond to these conditions. For that purpose, we integrate a generator of particles AGK 2000 (Palas® Model AGK 2000) purchased from Palas® society. A filtered air supply allow the reliability and reproducibility of the aerosol generation. The particle size spectrum of Palas® can be calibrated reproducibly in the range from approximately 5 nm up to 15 μ m, depending on the concentration of the solution and the pre-pressure of pressurized air. For this experiment, the microparticles chosen were SiC which presents different diameters less than or equal to 5 μ m.

E. Reference monitoring system

In order to verify the relevance of our system (impactor + SAW sensors), the tracking of the particles present in the chamber is compared with those obtained using an optical particle spectrometer for ambient PM monitoring Fidas® 100 purchased from Palas® society. Fidas® 100 are precisely developed for indoor quality and workplace measurements and furnish continuous and simultaneous measurements of PM1, PM2.5, PM4 and PM10.

II. MEASURE AND CHARACTERIZATION

A. SAW sensors response to SiC particles

Here, we present the evolution of the differential SAW sensor's phase signal throughout consecutive exposures to SiC particles. During the experiment, different microparticles concentrations are generated. The protocol is identical for each of them. When the desired concentration in the chamber of 1 m³ is reached, the aerosol generator is switched of and the pumping of the impactor is triggered to begin the air sampling. The measurements are acquired on both stages simultaneously. The

emanating aerosol contain particles in the [0, 10 μ m] range. Both stages consequently exhibit a phase variation. On the other hand, the optical spectrometer shows that the SiC dust obtained had a higher PM10 content than PM2.5. The PM10 stage exhibits, as expected, a higher phase shift in comparison to the PM2.5 stage, showing that more particles are larger than 2.5 μ m in the aerosol. Particles concentration injected, which varies in the 0 – 1600 μ g.m⁻³ range for PM10 and 0–120 μ g.m⁻³ for PM2.5.The deposition of particles on the sensitive area of the sensor give rise to a gravimetric effect that decrease the wave's phase velocity. That decrease in measured by monitoring the wave's phase at constant frequency. This variation is then compared with the particles concentration measured with the optical particle spectrometer is realized.

The gravimetric sensitivity of the SAW can by determined from the Sauerbrey equation (1) [5] (1):

$$S = \frac{df}{f0} \cdot \frac{A}{dm} \tag{1}$$

With f_0 the operating frequency, A the surface of the sensitive zone and dm and df the variations of mass and frequency respectively.

In the vicinity of the operating frequency, the phase is linear with the frequency. Therefore, the phase shift d ϕ is proportional to the mass variation dm (dm \propto d ϕ). The mass of particles collected on the surface is m = c × v with c the particle concentration and v the sampled aerosol volume. As v=Q.t where Q represents the sampling rate and t the sampling time, the mass variation is:

$$dm = c. Q. dt \tag{2}$$

Q remaining constant at 3 Lpm during the sampling and the concentration being considered stable during a small period dt, the particle concentrations can be calculated from (3).

$$c\alpha \frac{d\Phi}{dt}$$
 (3)

Figures 5 and 6 show the measurements obtained with our system and those from the reference optical method for both stages. We can see that both curves follow a similar shape. This similarity indicate a good correlation between the particle concentrations measured with the optical system and the phase variation of the SAW sensors.

The same protocol has been repeated with similar measurement on both stages, proving the repeatability of the measurements One can see figure 7 and 8, the phase derivatives represented as a function of the concentration measured with the optical method. By applying a linear fit of the data, the system's sensitivity predicted is 1.37×10^{-6} m°/s µg/m³ for PM10 and 6.9×10^{-6} m°/s µg/m³ for PM2.5.

The effect of successive measurements on the sensor lifetime has not been studied yet. Besides, an autonomous cleaning system is currently being considered.



Fig.5: Comparison between PM10 concentrations measured with the optical system Fidas® 100 (in blue) and SAW sensor phase derivative of PM10 stage in respect of time $d\phi/dt$ (in green).



Fig.6: Comparison between PM2.5 concentrations measured with the optical system Fidas® 100 (in blue) and SAW sensor phase derivative of PM2.5 stage in respect of time $d\phi/dt$ (in green).



Fig.7: Phase variation in terms of the particle's concentration for PM10.



Fig.8: Phase variation in terms of the particle's concentration for PM2.5.

B. Filtration characterization on the two stages

To check the ability of our system to target a precise size range, we took pictures of the sensors surface before and after the test using a digital microscope Keyence VHX-5000. On figures 9 and 10, we can observe the surfaces of the PM10 and PM2.5 stages respectively. It should be pointed out the inhomogeneities of the particle's distribution. The latter is linked to the surface of impactor nozzles.



Fig. 9: Appearance of the sensor's surface on the PM10 stage (zoom ×1000).



Fig.10: Appearance of the sensor's surface on the PM2.5 stage (zoom×1000).

III. CONCLUSION

In this work, we improved an original system for particulate matter concentration monitoring previously developed in our laboratory. This system integrates surface acoustic waves sensors and a custom-designed cascade impactor. This combination takes advantage of the SAW sensors high sensitivity to gravimetric effect, and the high selectivity of cascade impactors based on the aerodynamic diameter. Previous results have shown our ability to detect PM10 and PM2.5 from candles fumes. Here, we corroborate adaption of our device for atmospheric pollution monitoring with particles of SiC with different granulometries.

The integration of SAW sensors on the impaction plate allows real-time measurements. This feature represents a major improvement in comparison with the conventional cascade impactors. Besides, the use a differential configuration allows getting rid of operating conditions variations like pressure and temperature fluctuations. This is of great importance for performing outdoor measurements.

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REFERENCES

- Xing, Y. F., Xu, Y. H., Shi, M. H., & Lian, Y. X. (2016). "The impact of PM2.5 on the human respiratory system", Journal of thoracic disease, 8(1), E69-74.
- [2] L. Djoumi, M. Vanotti, V. Blondeau-Patissier, "Real time cascade impactor based on surface acoustic wave delay lines for PM10 and PM2.5 mass concentration measurement", Sensors, 18(1), (2018), pp. 255
- [3] P. DEFRANOULD and P. WRIGHT, "Filtres ondes de surface," vol. 2,pp. E2200–1.
- [4] D. Rabus, J.M. Friedt, S.Ballandras, G. Martin, E. Carry, V. Blondeau-Patissier, "High-sensitivity open-loop electronics for gravimetric acoustic-wave-based sensors", IEEE Trans. Ultrason. Ferroelectr. Freq. Control 60 (2013), pp. 1219-1226
- [5] Sauerbrey, G. Verwendung von Schwingquarzen zur Wägung dünner Schichten und zur Mikrowägung. Zeitschrift für Physik A Hadrons Nuclei 1959, 155, 206–222. (In German)