

Characterization and performance of an acoustic sensor for fission gas release characterization devoted to JHR environment measurements

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For over a decade, the IES laboratory has been working in collaboration with the CEA on the development of acoustic instrumentation in the nuclear field. As part of this collaboration, the IES acoustics team is developing a miniaturized gas composition sensor for in situ measurements of gas composition in a fuel rod. The first experimental use of an acoustic sensor to measure gas composition dates back to 2010 with the REMORA 3 experiment, which estimated the release of fission gas in an experimental fuel rod. Unlike the sensor tested during REMORA 3, this new sensor should be able to operate at 300°C, a performance made possible by the use of a piezoelectric element based on bismuth titanate (NBT). The material is screen-printed onto an alumina substrate. The manufacturing process and initial characterization were presented in the previous version of ANIMMA 2021 [1]. In this article, we will focus on the performance of this type of sensor by characterizing different gas mixtures representative of the gases inside a fuel rod, in order to estimate relative deviations under laboratory conditions.

Keywords —Acoustic sensor, composition, screen printing

I. INTRODUCTION

The research presented in this article is a direct follow-up to the work presented in 2021 at the ANIMMA conference [mettre ref]. This work detailed the study and design of an experimental gas composition sensor, dedicated to measurements in an experimental fuel rod of the Jules Horowitz Reactor (RJH). The laboratory developing these devices is the Institut d'Électronique et des Systèmes (Montpellier, France), and more specifically the acoustics team, which in part develops acoustic technologies dedicated to NDT operations in harsh environments. The sensor dedicated to measuring gas composition will be subjected to environmental constraints similar to those that might be encountered in the vessel of a PWR, namely:

- A maximum temperature of around 350°C
- A radiative flux of up to 1.175E13 (Thermal+Epithermal+Fast) [2].
- A volume of less than 3cm³

The sensor consists of one or more piezoelectric elements, which can operate in reflection or transmission mode. This piezoelectric element generates an acoustic wave in an acoustic cavity whose volume is of the order of 1 cm³. By measuring the time of flight of the acoustic wave, it is then possible to estimate the speed of the acoustic wave. The speed of the wave depends, among other things, on the atomic composition of the gas. The theoretical principle of sensor operation was demonstrated experimentally in 2010 in the OSIRIS reactor (CEA Saclay, Paris) by a sensor named CACP-1 [3], [4]. The principle of the measurement was to monitor gaseous releases from fuel pellets, which are either MOX or UO₂, during reactor operation. This experiment was carried out with a radiative flux similar to that which might be found in the vessel of a power generation reactor, but at a temperature of 150°C, a far cry from that envisaged for operation of the RJH. That's why, starting in 2015, our work focused on building a sensor capable of continuously measuring gas composition up to at least 350°C. To achieve this, we had to rethink the sensor manufacturing process and the materials used in its manufacture. We have abandoned lead zirconate titanate (PZT) in favor of bismuth niobate titanate (NBT), which has a Curie temperature of 650°C. And the piezoelectric element is no longer a solid material, but a layer of NBT deposited on an alumina ceramic substrate, which is relatively insensitive to oxidation and expansion compared with stainless steel, for instance. This manufacturing process avoids bonding problems and parallelism defects, as the adhesion between the substrate and the NBT is a direct bond.

The article will be divided into two parts, the first of which will deal with the principle of operation and manufacture of the sensor. The second part will focus on the sensor's initial results under laboratory conditions.

II. FABRICATION AND PRINCIPLE OF WORKING OF THE SENSOR

The innovative aspect of the sensor comes in part from the materials used in its manufacture and the manufacturing process [5]–[8]. The sensor is made exclusively of ceramic and is divided into two parts: The piezoelectric elements which are shown in Fig 1 A), these elements are disc-shaped and are assembled at the top and bottom of the cavity and bonded

with alumina cement. Fig 1 B) shows an assembled sensor with the gold connecting wires that enable the sensor to operate.

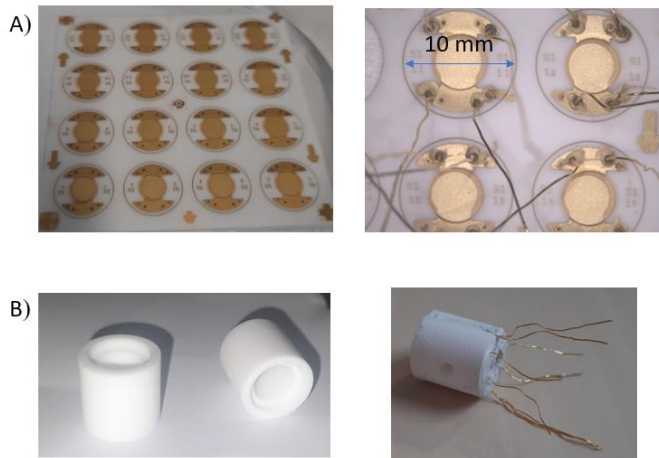


Fig 1: A) piezoelectric elements plate of 16 elements and a focus on 4 piezoelectric elements with gold wires soldering onto gold pads B) Acoustic cavity and sensor finalized with gold wire connectors

Piezoelectric elements are manufactured by successively depositing a first layer of a conductive material, in our case gold, on an alumina substrate. This is followed by a multitude of layers of the piezoelectric element, and finally a layer of gold to form the top electrode. This stack of layers is then fired at a temperature of 850°C to fuse the gold and sinter the ceramic, producing a ready-to-use piezoelectric element. The manufacturing method is similar to the low-temperature co-firing technique (LTCC). The layers are deposited using an ink that is a mixture of organic binder and the material to be deposited in fine powder form, in our case either gold for the electrodes or NBT for the active layer with piezoelectric properties. The ink is deposited *via* screen printing, a method that uses a doctor blade to force ink through a steel mesh stencil to reproduce the stencil pattern on the substrate. The sensor manufacturing process and the technical choices made to produce this sensor are discussed in more detail at the ANIMMA 2021 conference [1] and in this series of articles [5], [6], [8], [9].

The principle of the sensor is to measure the time-of-flight of an acoustic wave, and therefore to measure the wave velocity, and then, using an analytical model, to estimate the gas composition of the medium in which the wave is moving. The sensor can operate in two modes, the first using a single piezoelectric element and the second using two piezoelectric elements facing each other. If the sensor operates with a single piezoelectric element, it works in "reflection" mode, with the same piezoelectric element transmitting and receiving the wave. The second operating mode used for our sensor is the transmission mode, which uses two piezoelectric elements, one transmitting and the other receiving. This operating mode was chosen for several reasons:

- The use of two piezoelectric elements means that only one of the two elements can be electrically excited, which means that the second piezoelectric element is

more sensitive. This is impossible in reflection mode, as the same sensor emits and receives at the same time.

- When the sensor operates with two piezoelectric elements, the first echo received only travels one way into the cavity, so acoustic transmission losses are limited. On the other hand, in reflection mode the first echo necessarily makes one outward and one return travel.
- The advantage of having two piezoelectric elements is redundancy. In fact, there are two active elements per sensor, reducing the chance of global sensor failure.

The operating principle of the sensor is illustrated in Fig 2.



Fig 2 : Transmission sensors used for the experiments

In fact, the echoes emitted by the piezoelectric element are reflected several times in the resonant cavity. This phenomenon is shown on the tempograph in Fig 4, which is a measurement of the sensor. We can estimate the speed of sound by measuring the time between the various echoes, and knowing the height of the cavity. In a perfect gas, the acoustic wave velocity can be written as a function of the gas density and temperature [3]:

$$C = \sqrt{\frac{\gamma RT}{M}} \quad (1)$$

With:

γ : Laplace coefficient

R : Universal perfect gas constant

T : Gas temperature

M : gas molar mass

In the study of fission gas release, the gas present in the fuel rod can be assimilated to a diatomic gas mixture. The helium initially present in the fuel rod and the gaseous species produced, which are xenon and krypton. So the molar mass of the gas in the fuel rod can be written as the sum of the molar mass of the helium and the molar mass of the fission gases named M_{MFG} .

$$M = xM_{MFG} + (1 - x)M_{He} \quad (2)$$

The molar mass of M_{MFG} fission gases is a known ratio, depending exclusively on the type of fuel used. It's a mixture of xenon and krypton, in a proportion of $\frac{x_{Xe}}{x_{Kr}}=17$ [3].

At the first order, the equation of state does not take pressure into account when calculating velocity. By including Virial

equation of state at least to first order, it is possible to include pressure in the estimate. With β_{mix} and γ_{mix} related to Virial equation of state coefficient, and P the pressure of gas:

$$C^2 = \frac{\gamma RT}{xM_{MFG} + (1-x)M_{He}} (1 + \beta_{mix}P + \gamma_{mix}P^2 + \dots) \quad (3)$$

So, knowing all these parameters; the gas temperature being estimated by a thermocouple and the pressure being known by a manometer. we can estimate the ratio x which corresponds to the molar fraction of fission gases produced in the fuel rod.

III. EXPERIMENTAL MEASUREMENT

A. Material and method

The first tests are carried out at room temperature in a pressurized chamber, an image of which is shown in Fig 3. The sensor is placed inside, and sealed contacts are used to power the sensor and retrieve data. The sensor is connected to a TEKTRONICS TDS3032 oscilloscope and a Panametrics 5800 pulse (Dirac) generator. Two types of gas will be tested: firstly, pure helium, so as to calibrate the sensor and secondly, a mixture of helium, Xenon and Krypton, representative of the gas that might be found in a fuel rod.



Fig 3: Experimental pressure test bench

The sensor under test is a so-called transmission sensor with two piezoelectric elements facing each other as shown in Fig 2.

B. Gas characterization

First, the sensor is tested in pure helium at a pressure of 50 bar. The test chamber in which the sensor is housed is kept at a temperature of 25 degrees. Fig 4 shows two echograms of the same transducer, the different peaks representing multiple reflections inside the transducer's acoustic cavity. Two measurements are made, the first at 50 bar and the second at 25 bar. As the pressure is lower, echo attenuation is greater, as the medium is less dense. However, as long as two echoes are distinguishable from the noise, it is possible to measure the time of flight of the acoustic wave and thus obtain an estimation of the velocity.

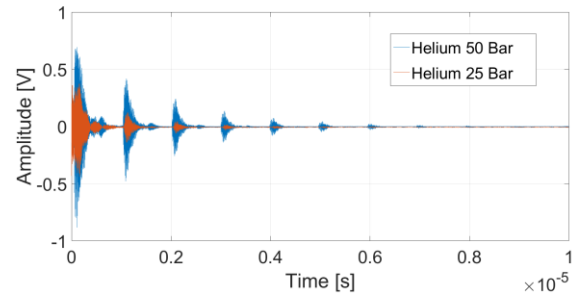


Fig 4: Echograms from the acoustic sensor tested at 50 and 25 bar in helium

For the estimation of the acoustic velocity, there is an uncertainty in the height of the acoustic cavity, which is machined to a height of 10 millimeters. But the machining uncertainties and the bonding process lead to uncertainties in its actual height, which is specific to each sensor. Given that we know the nature of the gas, which is helium, and its thermodynamic parameters (pressure and temperature), we can accurately determine the theoretical speed of sound in the gas from NIST data (Table I) [10];

SPEED OF SOUND (M/S) AT 25°C AND 50 BAR		Uncertainty
Data from NIST	1038.2	0.1%

For the sensor tested in this way, the real height of the cavity was estimated at $h=10.2$ mm; from the echogram it is possible to find the speed of sound in helium, which comes from the NIST data. Then Fig 5 shows the echogram of the sensor, this time in a mixture of helium, xenon and krypton, representative of the type of mixture that might be encountered when monitoring gas release in a fuel rod.

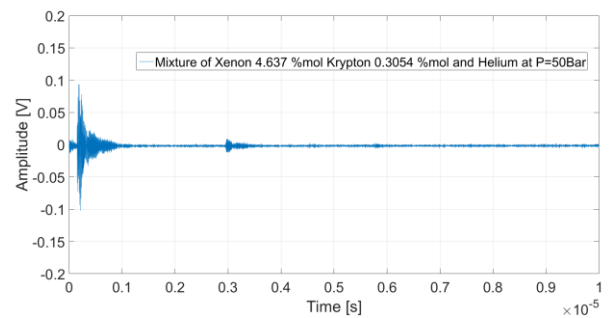


Fig 5: Echograms from the acoustic sensor tested at 50 for a mixture of Helium, Xenon and Krypton

The Table II shows the composition of the gas supplied by the manufacturer, together with its uncertainty. The same table shows the composition estimated by the Virial equation from the velocity measured by our sensor (Fig 5).

TABLE II
Comparison between gas composition estimated by the manufacturer and Sensor measurements

DATAS FROM GAS SUPPLIER (%MOL)		Uncertainty from gas supplier (%mol)	Measurement of the sensor (%mol)	Relative deviation for each gas (%)	
Xenon	4.637	Xenon	0.046	4.313	7.5
Krypton	0.305	Krypton	0.003	0.284	7.3
Helium	95.057	Helium	0.951	95.403	0.36

C. Discussion

Under laboratory conditions, initial measurements show a difference between the experimental and theoretical results, i.e. the manufacturer's data and our measurements, of between 7 and 8% (Table II). This accuracy is sufficient for our applications, but these measurements are carried out at room temperature and at a pressure of 50 bar, which gives a sufficient signal-to-noise ratio, as shown in Fig 5. It should be noted that the experimental rods in RJH will be at a temperature of around 300°C and a pressure of between 75 and 155 bar. The next stage of the tests is to combine the characterization of a gaseous mixture at a higher temperature, at least up to 300°C. According to the studies in [9], we should expect a decrease in acoustic performance. But at high temperature the pressure should be far more than 50 bar, we hope that this increase of pressure will compensate the diminution of acoustic performances.

IV. CONCLUSION

In this article, we have essentially presented the first experimental tests of our gas sensor. The materials used for the sensor's manufacturing make it an innovative sensor, unique at present in the state of the art. The sensor was able to characterize a mixture of three different gases with an accuracy of less than 10%, on the first sensor tested. Currently, a series of more than a dozen sensors similar to this one is being tested. And in the coming months, we plan to carry out further tests at a temperature of 300°C to validate the operation of our prototypes at high temperatures.

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