Validation of working of acoustic sensor for fission gas release characterization devoted to Jules Horowitz reactors in harsh condition up to 350°C and 120 bar

F. Baudry[1], G. Galeron[2], E. Rosenkrantz[1], P. Combette[1], PV. Bonzom[2] C. Destouches[2], F. Martin[2], D. You[3], R. Abadie[3], JY. Ferrandis[1*]

[1] IES, Univ. Montpellier, CNRS, Montpellier, France
[2] CEA-DES-IRESNE, Cadarache, Saint-Paul-Lez-Durance, France
[3] CEA-DES-ISAS, Université Paris-Saclay, Gif-sur-Yvette, France
(*) jean-yves.ferrandis@umontpellier.fr

Abstract— **For several years, the IES laboratory has been collaborating with the CEA to develop acoustic instrumentation for nuclear applications. As part of this collaboration, the IES acoustic team is designing a miniaturized sensor capable of measuring gas composition in situ within a fuel rod. Initial evidence of this approach dates back to 2010 with the REMORA 3 experiment, conducted in the OSIRIS reactor at CEA Saclay, which enabled acoustic measurements of fission gas release at 150 °C. The new sensors, however, are designed to operate up to 300 °C, thanks to a modified Bismuth Titanate (NBT) piezoelectric material, integrated by screen printing on an alumina substrate.**

This paper focuses on an experiment conducted in January 2024 in a high-pressure vessel at CEA Saclay, where the sensors were tested at 350 °C and 120 bar in various gas mixtures. Impedance and acoustic signal measurements were performed to evaluate sensor ageing and performance. After 79 hours, slight degradation was observed, but the sensor continued to emit acoustic signals and demonstrated its potential for reliable in situ gas composition monitoring. These results are a key step toward validating the device before irradiation tests planned for 2025.

Keywords — **Fission gas release, High temperature, Nuclear Instrumentation, Ultrasonic sensor**

I. INTRODUCTION

THE first experimental evidence of an acoustic measurement used to determine gas composition dates back to 2010 with the REMORA 3 experiment, which measured the release of fission gases from fissile fuel. This experiment was carried out in the OSIRIS reactor located at CEA Saclay in France. The setup allowed the devices to be tested under irradiation at 150 °C [1], [2], corresponding to the maximum operating Curie temperature permitted by PZT elements.

However, the new sensors presented in this paper are designed to operate up to 300,ÄØ¬∞C. This improved performance is made possible through the use of a modified Bismuth Titanate piezoelectric material (NBT). The material is integrated using a screen-printing process on an alumina substrate [1], [3]. The fabrication process and initial characterizations were previously presented at ANIMMA 2021 [4] and ANIMMA 2023 [5], [6].

The research presented here is a direct continuation of the work introduced at the 2023 ANIMMA conference, which detailed the design of an experimental gas composition sensor for measurements in an experimental fuel rod of the Jules Horowitz Reactor (RJH).

These devices are developed by the Electronic and System Institute (IES) in Montpellier, France, specifically by the acoustics team, which also develops acoustic technologies for NDT applications in harsh environments. This sensor is intended to monitor fission gas release in experimental fuel rods. Its operational requirements are as follows:

The sensor must remain functional at temperatures of at least 350 °C

It must perform measurements at pressures of at least 60 bar

The sensor operates based on an acoustic time-of-flight measurement. An analytical model is then used to estimate the gas composition as a function of temperature, pressure, and acoustic velocity.

The manufacturing process and operating principles of the sensor were described in detail in [4]. At IES, we tested the sensor's behavior under varying pressures at room temperature [6], and these tests validated the concept of a gas composition sensor.

This article presents experiments conducted at the DRMP laboratory at the CEA Saclay site, where the sensor is tested in an autoclave at temperatures up to 350 ° C and pressures up to 125 bar.

In the first part, we will describe the sensor preparation process, followed by the experimental setup and test protocol. Finally, the third section will present the experimental measurements and results.

II. PRESENTATION OF THE SENSOR

The sensor tested in this study was manufactured and assembled at IES, as shown in Fig. 1, and is composed of two main components. The first is a piezoelectric, or active, element that emits the acoustic wave. As shown in Fig. 1, each sensor consists of two piezoelectric elements positioned face-to-face. The second component is a cylindrical ceramic cavity. All components are assembled using alumina cement, an adhesive capable of withstanding thermal stresses up to 1000 °C in theory. The piezoelectric ceramic used is based on bismuth titanium niobate (NBT), a material extensively studied in [7]. Its Curie temperature, depending on the specific application, is approximately 640 °C.

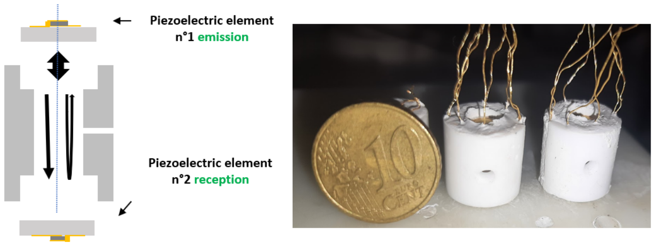

Fig 1. Transmission sensors used for the experiments

The sensor has four connection channels, two for each acoustic element. However, due to technical constraints of the experiment, the sensor had to be connected using only three wires, meaning that both piezoelectric elements share a common ground. Fig. 2 shows the sensor prepared for testing, with wire extensions. These extensions allowed for improved connectivity between the sealed electrical contacts and the sensor.

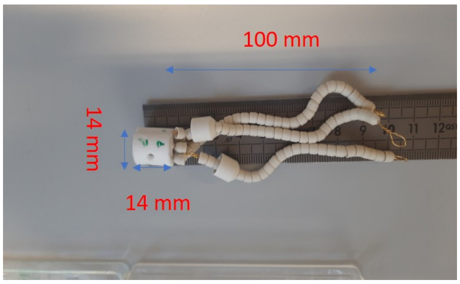

Fig 2. Gas sensor ready to be tested and integrated into the autoclave

Now we're going to go into detail about the experiment and the experimental protocol.

III. EXPERIMENTAL SET-UP AND MEASUREMENTS

The aim of the experiments was to characterize the sensor under thermal, pressure, and gas mixture conditions similar to those found in an experimental fuel rod. These conditions can be reproduced in an autoclave.

A. Material and Methods

The autoclave at DRMP is a device capable of containing a gas mixture at temperatures up to 350 °C and pressures up to 120 bar. A schematic view of the autoclave is shown in Fig. 3. It consists of two sections: a cold section and a hot section. The cold section houses the pressure gauges, gas mixture cylinders, and safety devices. It is connected to the hot section via a pipe, and it is not possible to isolate the two sections from each other. The hot section is equipped with a temperature-controlled heating collar, which raises the chamber to the desired temperature. A type K thermocouple placed inside the chamber is used to measure the temperature.

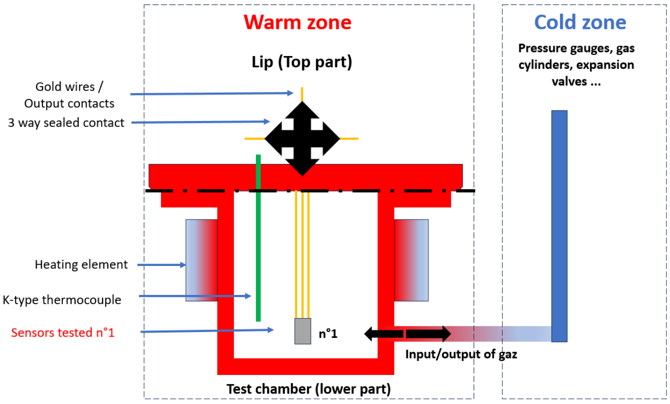

Fig 3. Schematic cut view of the autoclave

The sealed contacts allowed us to connect our instrumentation and monitor the acoustic sensors. The sensor is integrated into the autoclave in three stages, as shown in Fig. 4. It is connected to the sealed contacts via pure gold wires with a diameter of 300 µm. Additionally, the sensor is suspended to avoid any mechanical coupling with the internal wall of the chamber.

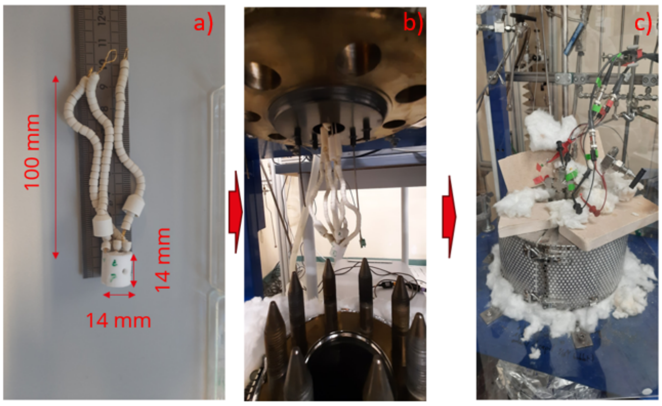

Fig 4. (a) Presentation of the sensor, (b) Sensor before closing the autoclave, (c) Autoclave closed and insulated ready to operate with the instrumentation connected

Once the autoclave is closed and sealed, a number of preparatory steps are required to ensure it is leak-tight. The autoclave then undergoes a helium purging process, which consists of increasing the pressure, then reducing it, and repeating this cycle three times. This process results in a helium concentration of over 99.9% within the autoclave.

For this experiment, two different acquisition systems were used. The first is dedicated to acquiring acoustic data, generally in the form of echograms, as shown in Fig. 5a. From these echograms, the acoustic velocity in the gas can be estimated, which in turn allows for an estimation of the gas composition.

This acquisition chain consists of a Tektronix TDS 3032 oscilloscope and an Olympus Panametrics 5800 acoustic signal conditioner, with data acquisition performed on a laptop computer running dedicated software. The second acquisition system uses a Keysight E4990A impedance analyzer, which enables monitoring of impedance changes over time and as a function of temperature (Fig. 5b).

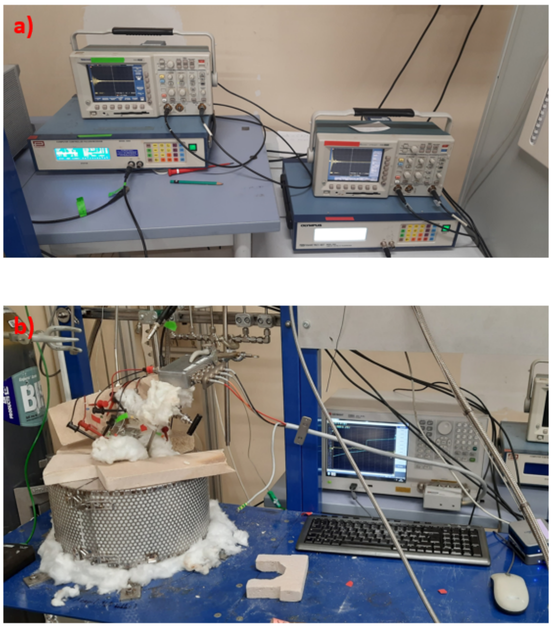

Fig 5 : (a) Photograph of the acoustic test benches used to measure velocity, (b) Impedance analyser Keysight E4990A used to measure real and imaginary impedance

B. *Experimental environment*

The experiment was conducted over a period of four days, during which various gas mixture compositions were evaluated. The experiments presented in this article were performed at distinct temperature levels: 30, 100, 250, and 350°C. These conditions are further illustrated by the impedance measurements displayed in Fig. 6. The gas utilized was sourced from a Messer Group cylinder, consisting of 23.6% xenon, 2.04% krypton, and the balance helium. Hereafter, this will be referred to as the He-Xe-Kr mixture.

Subsequently, gas composition measurements will be conducted at 250°C and 350°C under isothermal conditions.

IV. RESULTS AND DISCUSSION

A. *Impedance vs. Time*

To study the sensor's evolution, its impedance was measured in situ throughout the experiment. Measurements were conducted at various temperatures, ranging from 30°C to 350°C. The sensor consists of two opposing piezoelectric elements, designated as number 1 and number 2 in Fig. 1. Their temperature-dependent impedances are presented in Fig. 6. The chosen frequency window enables the observation of the first two impedance peaks (approximately 5 MHz and 9 MHz). These peaks are investigated due to the enhanced efficiency of the [1; 10] MHz bandwidth for acoustic emission in gases.

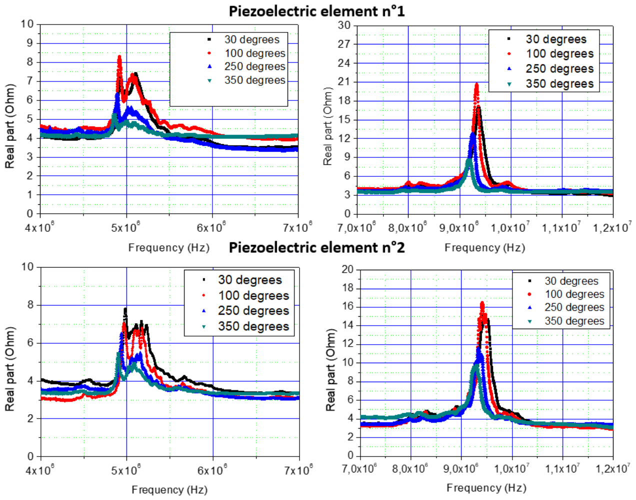

Fig 6. Measurement of real parts of electrical impedance as a function of temperature between [4; 7] MHz and [7; 12] MHz of piezoelectric elements number 1 and number 2

The measurements indicate that the piezoelectric elements responses change with temperature. This observed decrease is linked to a reduction in the coupling coefficient parameter kt, but it does not indicate a degradation of the piezoelectric elements. This phenomenon has been consistently observed in previous laboratory experiments, and post-test analysis confirmed that the acoustic elements suffered no degradation [6].

B. *Measurement at 250 degrees in He-Xe-Kr mixture*

In this experiment, the autoclave was held at a constant temperature of 250°C and a pressure of 124 bar. Initially, an He-Xe-Kr mixture, comprising 10% xenon and 0.9% krypton, was injected into the chamber. Subsequently, helium was introduced into the autoclave over an 11-hour period to progressively decrease the xenon concentration. To ensure isobaric conditions were maintained, a deliberate leak was created to offset the helium injection. Fig. 7 presents the velocity measurements recorded throughout the experiment, demonstrating the gradual convergence of the echoes over time.

Furthermore, at the experiment's onset, only two echoes are visible. However, after 10 hours, a multitude of echoes appear with increased amplitudes. Experimentally, we found that decreasing the xenon concentration reduces attenuation. Fig. 8 displays the velocity measurements extracted from the echograms between 7 PM and 6 AM. After 10 hours, the velocity measured in the autoclave approaches the speed of sound of helium at 124 bar and 250°C, which is 1382 m/s (data from the NIST institute).

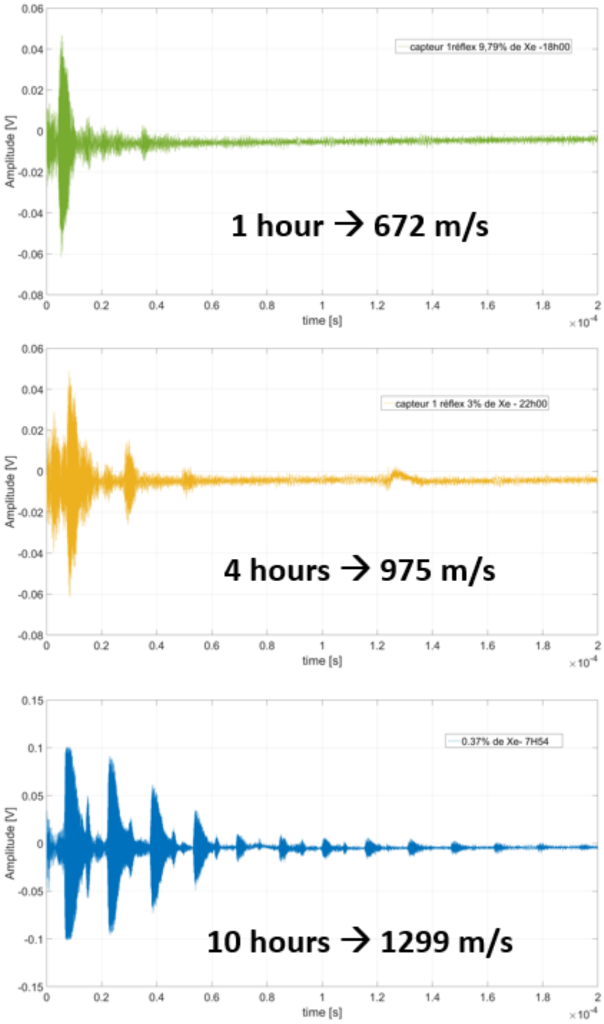

Fig 7. Three echogram measurements taken at 1 hour, 4 hours and 10 hours respectively

As mentioned previously, by measuring acoustic velocity, temperature, and pressure, we can estimate the xenon composition. This estimation has been detailed in several articles [1, 2] and is based on the following equation:

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

with C the acoustic velocity, T the temperature and P the pressure, M_{Xe} and M_{He} the molar masses of the different gases, R the perfect gas constant, γ which in our case is equal to 5/3 and β_{mix} and γ_{mix}, the virial coefficients corresponding to the gas. From these parameters, it is then possible to trace the x parameter, which is the mole fraction of xenon. These parameters can then be used to determine the x parameter, which is the mole fraction of xenon. Using the measurements from Fig. 8 and Equation (1), an estimate of the xenon composition is provided in Fig. 9.

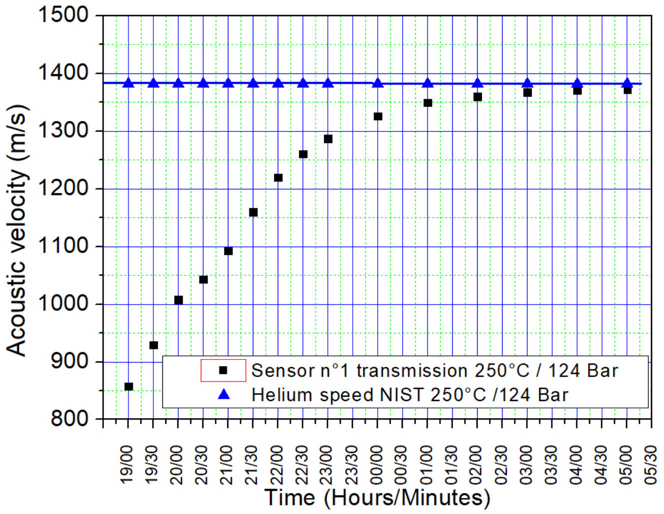

Fig 8.: Acoustic speed measurements between 7 PM and 5:30 AM, i.e. for 10 hours and 30 minutes

Uncertainties in xenon composition are directly linked to uncertainties in temperature measurements. This demonstrates the sensor's ability to track variations in gas composition at 250°C. However, these measurements are estimations, and no mass spectrometry was performed.

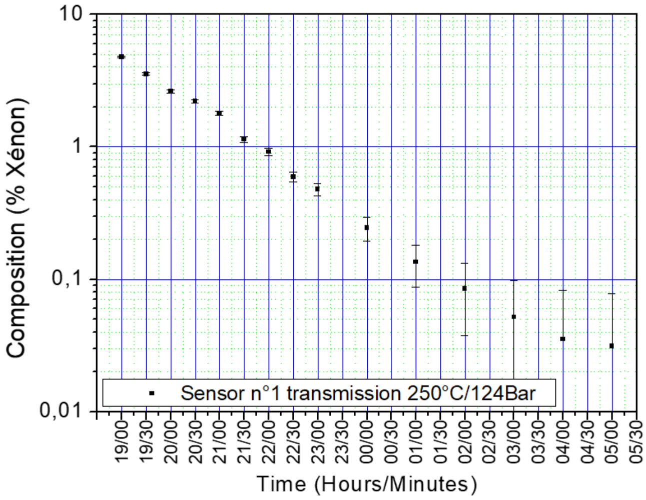

Fig 9. Composition measurements between 7 PM and 5:30 AM, i.e. for 10 hours and 30 minutes

C. *Measurement at 350°C in pure He*

The autoclave is now at a temperature of 350°C, containing a helium mixture of over 99%. The advantage of this mixture is its low attenuation, which allows us to test the minimum operational pressure. Fig. 10 shows three echograms for three different pressure values: 119 bar, 90 bar, and 60 bar.

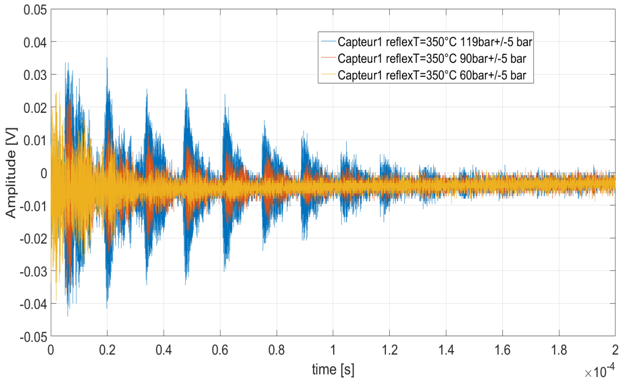

Fig 10. Measurements of the sensor in helium at 350°C at different pressures

We found that the sensor was operational at 350°C and successfully performed an acoustic measurement. This demonstrates that both requirements have been met: first, operating at 350°C, and second, being able to measure at a pressure of at least 60 bar.

V. CONCLUSIONS

The gas composition sensor developed by IES in partnership with CEA IRESNE successfully performed an initial measurement of changes in gas composition. The temperature and pressure conditions, 250°C and 124 bar respectively, were representative of experimental conditions in Material Testing Reactor experiments. We studied concentrations ranging from 10% xenon to less than 1%, and all sensor components withstood the thermal stresses. The temperature was then increased to 350°C, and we tested a mixture with a low helium content. Encouragingly, many echoes are visible at pressures between 120 and 60 bar.

Post-test analysis confirmed that the acoustic elements suffered no degradation. The sensors remained operational after a total duration of 34 hours at 250°C and 10 hours at 350°C. The radiation reliability of the final sensor remains to be tested.

These encouraging results suggest the sensor's potential application as a fission gas release characterization device within the test loops of the forthcoming RJH reactor, particularly the Madison loop.

ACKNOWLEDGMENT

This work was financed by a partnership between IES and the CEA IRESNE institute and more specifically the LISM laboratory and the INSNU project. The authors would like to thank the CTM (Montpellier Micro and nanoelectronics Technology Centre). Finally, we would like to warmly thank Vincent Lechien (technician at Montpellier University) for his support on screen printing methods.

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