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Application of Cavity Ring Down Spectroscopy in the monitoring of gases resulting from failures innuclear fuels

Matos, P.*; Landulfo, E.; Peixoto, G.; Pompei, G. and Raele, M.P.

Nuclear and Energy Research Institute, IPEN-CNEN/SP, Av. Prof. Lineu Prestes, 2242
Cidade Universitária, São Paulo, SP, Brazil.
**Priscila.m@ipen.br*

1. Introduction

Energy production is currently undergoing a period of reassessment due to the environmental and climate impacts arising from the use of fossil fuels. On the other hand, it is expected that the consumption of electricity will double every 30 years[1], demonstrating that just replacing the current sources will not solve the problem. In this scenario, nuclear energy emerges again as a solution for the generation of electric power[2] [1], however, its feasibility depends on the implementation of adequate safety measures for the operation of the reactors.

The integrity of fuel elements (FE) is crucial to ensure the safety and proper functioning of nuclear reactors. The release of gases that are fission products, such as Kr and Xe, is indicative of FE cracks.

One of the techniques used to monitor gases resulting from failures in nuclear fuels is differential optical absorption spectroscopy, gas chromatography, and fluorescence spectroscopy[3].

Cavity Ring Down Spectroscopy (CRDS)[4] is also based on optical absorption by molecules/atoms in a gaseous or liquid sample, i.e., it measures extremely low concentrations of trace gases, and is useful for monitoring gases emitted by failures in nuclear fuels, such as parts per million (ppm) to parts per billion (ppb)[5]. As shown in Figure 1 below, this technique is achieved by creating a high-precision optical cavity with length L, formed by highly reflective plano-concave mirrors (R_1 and R_2) that trap the incident light, thus enabling multiple reflections within the cavity.

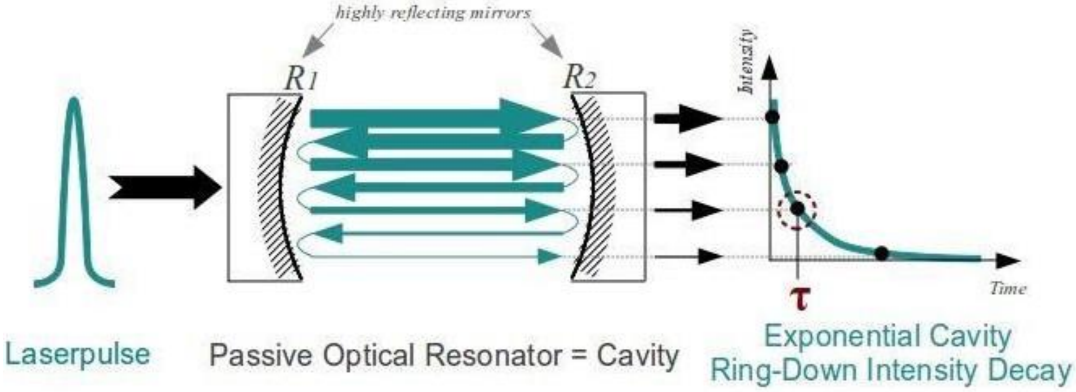

Figure 1: Schematic diagram explaining the principle of CRDS. The arrows present in the cavity indicate the round-trip trajectory of the light, losing intensity after each pass.
Source: Layertec[6].

Despite this relevance, the literature lacks reports on CRDS applications specifically aimed at identifying failures in the FE of PWR (Pressurized Water Reactor) reactors or research reactors. This information gap underscores the need to explore more specialized detection techniques to assess the integrity of FE in these specific contexts.

In the context of PWR-type nuclear reactors, the integrity of the fuel rod plays a crucial role, as it not only contributes to the correct functioning of the reactor, but also plays an essential role in the containment of nuclear fuel. The fuel rod, being the first barrier against the release of fission products, is of fundamental importance to ensure the overall safety of these reactors [7].

Therefore, the research and development of advanced methods, such as the use of CRDS, aimed at detecting specific failures in the FE of PWR reactors, are imperative to strengthen the structural integrity of these elements, thus contributing to the continuous and effective safety of nuclear operations.

2. Objective

The present study aims to propose a system for the detection of gaseous fission products via CRDS and to implement a benchtop system for gas detection.

3. Materials and methods

To this end, an extensive search of existing databases was conducted, including WebofScience, Scopus, Google Scholar, etc., to better understand the possibilities and obstacles.

A study was carried out on the potential elements of interest for the detection of failures in EC, some examples are Xenon (Xe), Krypton (Kr), Promethium (Pm), Technetium (Tc), Strontium (Sr) among other candidates that are elements resulting from the division of nuclei in nuclear reactor reactions.

A survey was also carried out in the spectroscopic database of the NIST (National Institute of Standards and Technology)[8] in relation to the absorption lines of the selected elements, considering intensity and possible interferences in their measurement.

Additionally, a benchtop CRDS system will be implemented, operating in an optical region close to one of the selected spectral lines. In this step, a CRDS cavity will be assembled and aligned, with mirrors that meet the specifications of the previous steps. A detection system (spectrometer) will be coupled via fiber optics as well as a broadband light source.

4. Results and Discussion

Among the gaseous fission products {Xenon (Xe), Krypton (Kr), Promethium (Pm), Technetium (Tc) and Strontium (Sr)}, we limit transitions of interest at wavelengths compatible with widely available and low-cost laser sources. These sources are based on active Nd:YAG and He:Ne media, with emissions corresponding to 355 nm, 532 nm, and 632 nm, respectively. The 1064 nm transition was ruled out due to the complexity of the detectors in the infrared range. The selected lines are close to the emissions of the lasers, which makes them more accessible and less expensive, since the lasers are widely produced and available. Taking into account the sources available at the Center for Lasers and Applications (CELAP) and the associated costs, we chose to limit our measurements to the 355 nm, 532 nm, and 632 nm regions.

Among the fission product options for fault monitoring in Table I below, we have selected the following elements and lines with high intensity (>100).

Table I – Survey of the NIST database in relation to the absorption lines of the selected elements.

Element	Observed Wavelenght Air (nm)	Relativity intensity
Tc I	353,5828	800
Tc I	353,8678	800
Tc I	356,0319	800
Tc I	356,8853	800
Tc I	532,0198	600
Sr I	532,9813	500
Xe I	631,80620	500
Pm I	632,384	700
Pm I	632,384	700

Source: NIST.

Xenon is interesting in view of the 532nm line that can be accessed with a widely commercially available laser.

5. Infrastructure

To conduct this experiment, we have a resonant optical cavity with a length of approximately 1 m.

In addition, we have at our disposal plano-concave mirrors that will be positioned at the ends of the selected cavity.

The experiment will involve the use of a Licel spectrometer, capable of simultaneously detecting several wavelengths, with high temporal resolution.

As a light source we have a commercial supercontinuous source (*supercontinuum* – SC) that has a wide bandwidth (supplied by Leukos, with a frequency of 8 kHz). This source has an approximate temporal duration of 1 ns, and emits wavelengths ranging from 200 to 2400 nm with an average power of 200 mW.

To facilitate the evacuation of the given cavity, we will employ a chemical vacuum pump.

6. Next steps

Let's start by acquiring the mirrors and assembling a vacuum cavity. Subsequently, we will introduce a known gas to start the system running. We will make comparisons between the vacuum cavity and the one containing a gas that will simulate the lines of the candidate element.

Through pressure we will be able to quantify the sensitivity of the system and see its compatibility with the proposed application.

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