

## Towards a real-time quantification of aerosol deposit on walls during fires in the nuclear industry

Kort A \*, Plumecocq W \*\*, Mastori H \*\*, Pailloux G \*, Wiszniewski P \*, Lakhmi R \*\*\*

\*Autorité de Sûreté Nucléaire et de Radioprotection (ASN), PSN-RES/SCA, Saclay, F-91400, France

\*\*Autorité de Sûreté Nucléaire et de Radioprotection (ASN), PSN-RES/SA2I, Saint-Paul-lès-Durance, F-13108, France

\*\*\* Mines Saint-Etienne, Univ Lyon, CNRS, UMR 5307 LGF, Centre SPIN, Saint-Etienne, F-42023, France

### Abstract:

This work has been carried within a joint research project Orano/ASN. It focuses on studying aerosol deposition on fire rooms walls, a crucial aspect for enhancing fire safety measures and radiological risk management in nuclear fuel reprocessing facilities. To this end, a multi-sensor dynamic device was developed for real-time aerosol deposition measurement, along with an expert system designed to predict the fraction of radionuclides, in the form of aerosols, that remain undeposited on the fire room walls. The multi-sensor system includes a resistive sensor and a transmissive NDIR (*Non-Dispersive InfraRed*) CO<sub>2</sub> sensor, which measures the CO<sub>2</sub> released during the regeneration of aerosols deposited on the resistive sensor. First, the electrical response of the resistive sensor was characterized, and a polarization voltage of 10 V was chosen for real-time measurements. Finally, an Artificial Intelligence (AI) expert system was developed based on a database of 7 million SYLVIA (software to study ventilation network, fire, and airborne contamination) calculations, to predict the fraction of radionuclides not deposited on the fire room walls. SYLVIA was validated by quantitative experimental data.

### 1 INTRODUCTION

PUREX process (*Plutonium Uranium Refining by Extraction*) is used to recycle spent nuclear fuel by separating valuable materials, mainly uranium and plutonium, from radioactive waste. In this process, spent fuel is dissolved in an aqueous nitric acid solution, resulting in a mixture containing actinides and fission products in various oxidation states. Uranium (+VI) and plutonium (+IV) are selectively extracted from the acidic aqueous phase into an organic phase composed of TBP (*Tributyl Phosphate*) diluted in HTP (*Hydrogenated TetraPropylene*), due to TBP's strong affinity for these high oxidation states. However, the organic phase, consisting of TBP and HTP, is highly flammable (HTP has a flash point of 52°C), presenting a fire hazard involving radioactive materials and potentially resulting in the release of radioactive and toxic substances into the environment.

The total activity concentration corresponding to this release, referred to as the “fire source term” S (Bq), serves as input data for assessing the radiological consequences of a fire. Within a joint research project Orano/ASN, the ASN's methodology for estimating the fire source term is based on the estimation of four coefficients and is expressed as follows:

$$S = \sum_{i=1}^n \sum_{j=1}^m S_{ij} = \sum_{i=1}^n \sum_{j=1}^m A_{t,ij} \cdot C_{0,ij} \cdot C_{1,ij} \cdot C_{2,ij} \cdot C_{3,ij}$$

- $S_{ij}$  : Activity of radionuclide  $i$ , in physico-chemical form  $j$ , released into the environment [Bq],
- $A_{t,ij}$  : Total activity of radionuclide  $i$ , in physico-chemical form  $j$ , present in the room(s) affected by the fire [Bq],
- $C_{0,ij}$  : Ratio of activity of radionuclide  $i$ , in physico-chemical form  $j$ , involved (or releasable) in the fire [-],
- $C_{1,ij}$  : Airborne release fraction of radionuclide  $i$ , in physico-chemical form  $j$  [-],
- $C_{2,ij}$  : Fraction of aerosols of radionuclide  $i$ , in physico-chemical form  $j$ , not deposited on the fire room walls [-],

- $C_{3,ij}$  : Fraction of aerosols of radionuclide i, in physico-chemical form j, not deposited in the exhaust pathways [-].

These coefficients ( $C_0$  to  $C_3$ ) describe the various mechanisms governing the emission and transfer of radioactive materials from the source to the environment during fires. To improve fire safety and radiological risk management in nuclear fuel recycling facilities, it is necessary to characterize each of these coefficients. This work focuses on the  $C_2$  coefficient, which relates to aerosol deposition in rooms (1- $C_2$ ) during fire events. Reliable data on aerosol deposition require the development of suitable metrological and numerical tools. To this end, an innovative dynamic multi-sensor device has been developed for real-time aerosol deposition measurement, along with an AI expert system designed to predict the fraction of radionuclides, in aerosol form, not deposited on room walls. This paper presents the qualification of the resistive sensor used in the device, the dynamic measurement system, and the AI expert system based on a database of 7 million SYLVIA calculations.

## 2 EXPERIMENTAL APPROACH FOR ESTIMATING THE 1-C2 COEFFICIENT

The first series of tests aims to quantify aerosol particles deposits produced from the combustion of 30%<sub>v</sub> TBP and 70%<sub>v</sub> HTP fuel mixture, representative of solution ratios used in nuclear fuel reprocessing facilities. For this purpose, a resistive sensor technology, initially developed for automotive emissions, and an NDIR CO<sub>2</sub> sensor were selected.

The resistive sensor operates based on the conductive properties of aerosols. Its sensitive element consists of interdigitated electrodes, optically engraved and separated by 20 µm, on a platinum screen-printed onto an alumina substrate. The resistive sensor measures change in conductance between the interdigitated electrodes caused by aerosol deposition when a polarization voltage is applied. Without aerosol, the measured conductance corresponds to that of the alumina substrate and is close to zero. As aerosols deposit, the sensor's conductance increases, allowing qualitative monitoring of the deposited aerosol mass. Since the sensor's principle depends on the aerosol's electrical properties and its response is influenced by factors such as polarization voltage, aerosol composition, and spatial position within the flow, its response must be studied specifically for the TBP/HTP mixture.

Above a certain deposited mass, the sensor's sensitive surface must be regenerated by burning off the aerosol using a screen-printed heating resistance located on its rear face, operating at 600 to 700°C. This regeneration phase enables quantification of the deposited particle mass by measuring the CO<sub>2</sub> concentration emitted during this phase.

### 2.1 Evaluation of the electrical response of the resistive sensor for the TBP/HTP mixture

The resistive sensor is exposed to aerosol produced by the combustion of the TBP/HTP mixture. The experimental test bench is shown in *Figure 1 (a)*. It consists of a combustion chamber where the fuel mixture is placed and ignited under controlled conditions, with air (oxidizing gas) injected to maintain an oxygen concentration of 21%. The combustion chamber is topped by a vertical column that serves as a transport pathway for the generated aerosols. Both the chamber and the column are thermally insulated, and the column is heated to 150°C to prevent aerosol deposition by thermophoresis.

At the top of the column, a fume hood ensures safe evacuation of gases, and sampling ports allow for aerosol collection and connection of metrological equipment. One of the outlets is connected to a 5 cm diameter glass tube, where aerosols are conditioned in a tube furnace also maintained at 150°C. This temperature simulates conditions close to those observed on room walls during large-scale fire scenarios, allowing realistic sensor evaluation. The resistive sensor is positioned inside the glass tube, at the outlet of the furnace. Particle size distribution is measured using a SMPS (*Scanning Mobility Particle Sizer*) located downstream of two Palas VKL10 dilution systems. The conductance signal from the resistive sensor is recorded by a Keithley 6517A multimeter, operated via a Python-controlled acquisition system.

*Figure 1 (b1) and (b2)* shows TEM (*Transmission Electron Microscopy*) images of TBP/HTP aggregates, highlighting the formation of fractal-like structures. These aggregates may carry an adsorbed phase of phosphoric acid *Figure 1 (b2)* [1], commonly generated during combustion of TBP/HTP mixtures.

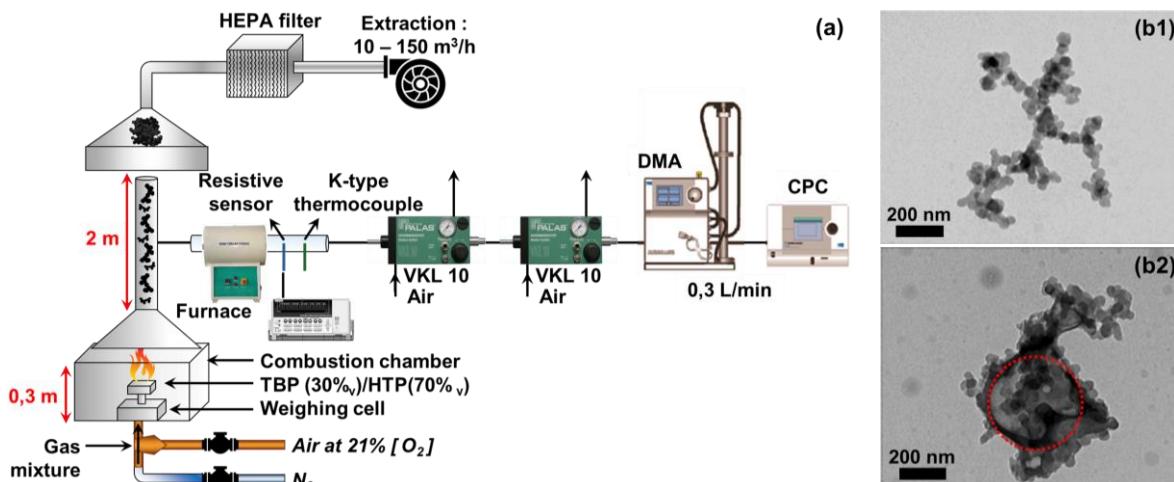


Figure 1: (a) Experimental setup for aerosol generation and deposition. TEM images of a TBP/HTP aggregate: (b1) without adsorbed phase, (b2) with adsorbed phase [1]

Figure 2 presents two complementary analyses: Figure 2 (a) shows particle size distribution at different times, while Figure 2 (b) illustrates the evolution of the MLR (Mass Loss Rate) throughout the test. Figure 2 (a) reveals that the aerosol particle size distribution evolves as the solvent burns. Upon ignition at  $t_0=0$  s, a mode centered around 20–30 nm with a concentration of about  $5 \times 10^7$  particles/cm<sup>3</sup> appears, corresponding to the formation of primary ultrafine aerosols. 3 minutes after ignition, the distribution becomes monomodal and remains stable until around 18 minutes, with a median diameter of  $216 \pm 15$  nm, a geometric standard deviation of  $1.5 \pm 0.1$ , and a number concentration of  $4 \times 10^7 \pm 10\%$  particles/cm<sup>3</sup> (corrected for a dilution factor of 100). This period corresponds to a quasi-steady combustion phase, during which particle size and concentration vary slightly, indicating stable aerosol production without strong thermal dynamics. This steady phase is also observed in Figure 2 (b), where the MLR rises quickly after ignition, reaching a plateau around 70 g/min, and then remains nearly constant. Around 21 minutes, just before flame extinction, both figures highlight significant changes. Figure 2 (a) shows an increase in particle concentration from  $3.5 \times 10^7$  to  $4.5 \times 10^7$  particles/cm<sup>3</sup>, accompanied by a reduction in particle size and the emergence of a new mode around 174 nm. Simultaneously, Figure 2 (b) indicates a brief drop in MLR followed by a sharp peak reaching 80 g/min. These changes coincide with the onset of intense boiling and the thermal degradation of TBP, which likely contributes to the increase in particle number by releasing additional volatile compounds that nucleate or condense into smaller particles under high temperature and turbulence conditions. This parallel behavior provides a clearer and more comprehensive measure of aerosol dynamics during combustion. After extinction ( $t_0+24$  min), sensor regeneration occurs ( $t_0+27$  min), which is visible through a new mode centered at 35 nm, indicating renewal of the aerosol layer on the sensor's sensitive face.

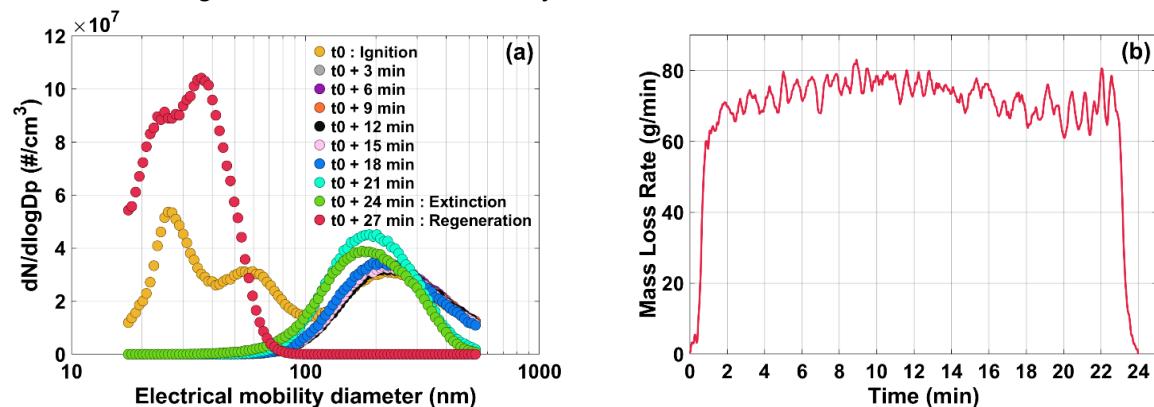


Figure 2: (a) Evolution of the aerosol size distribution produced by a TBP/HTP mixture, from flame ignition to sensor regeneration. (b) Mass loss rate (MLR) as a function of time during the same experiment

To study the effect of polarization voltage on the resistive sensor response, the same sensor was exposed to the aerosol previously described under identical conditions at two polarization voltages: 0.1 V (*Figure 3 (a)*) and 10 V (*Figure 3 (b)*). The conductance was recorded until the flame extinction. Three successive exposure cycles were performed, with sensor regeneration between each cycle.

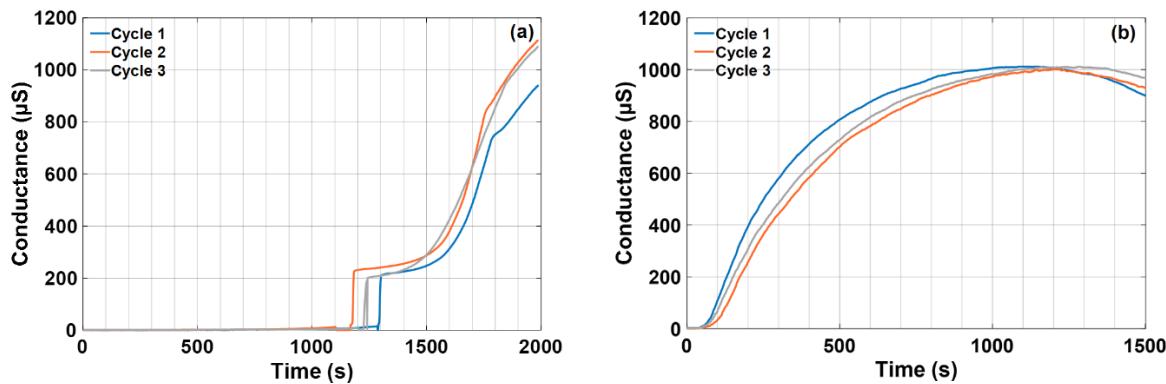


Figure 3: Evolution of conductance versus exposure time (time origin set at the end of the regeneration phase) for different bias voltages: (a) 0.1 V and (b) 10 V, over three successive tests

Two regimes are identified: an “intermediate” regime at 0.1 V and a fully “dendritic” regime at 10 V, as reported in previous studies [2].

At 0.1 V, conductance shows an initial sharp increase followed by a plateau. In this regime, electrophoresis forces are weak, preventing aerosol particles from aligning and forming parallel dendritic bridges between electrodes. As a result, the deposit is poorly structured, and the junctions between electrodes form discontinuously. After the plateau, conductance gradually increases again.

At 10 V, conductance rises continuously, without abrupt jumps. In this case, stronger electrophoresis forces are more influential on deposit structuring, enhancing the formation of multiple dendrites growing in parallel, with successive bridge formation between electrodes. At this voltage, conductance eventually saturates, then decreases continuously. This decrease can be associated to physico-chemical changes in the aerosol caused by Joule heating at the sensor surface under 10 V. Based on these conclusions, a polarization voltage of 10 V will be applied for the upcoming real-scale experiments.

## 2.2 Quantification of the deposited mass

Once the sensitive face of the resistive sensor is contaminated, quantification is performed by measuring the CO<sub>2</sub> emitted during aerosol regeneration [3]. For the upcoming large-scale fire tests planned in 2027 in PLUTON facility (400 m<sup>3</sup> volume, ASNR Cadarache), a new dynamic quantification system enables online monitoring (*Figure 4*). This system involves positioning the resistive sensor on the wall surface to be contaminated by aerosol particles while recording the electrical response. The sensor is then isolated inside a regeneration cell via a pneumatic valve, allowing quantification of the CO<sub>2</sub> emitted during aerosol regeneration with a NDIR CO<sub>2</sub> sensor. The experimental data will be used to improve the expert system's predictions.

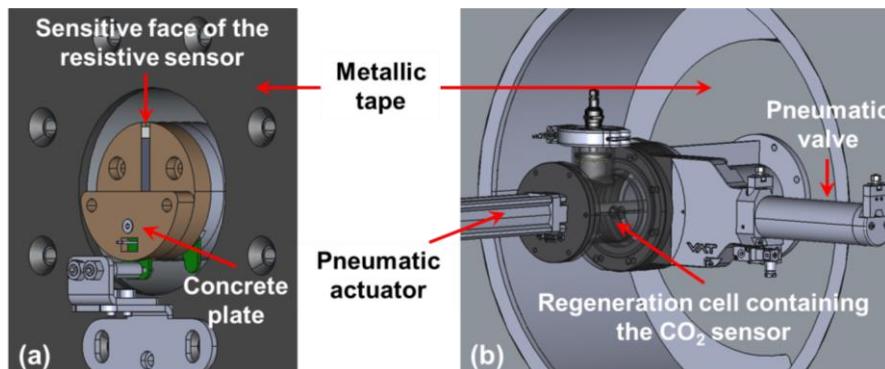


Figure 4: 3D view of the new dynamic quantification system: (a) front view of the wall showing the sensor in measurement position; (b) rear view of the wall showing the regeneration cell

### 3 THE EXPERT SYSTEM FOR ESTIMATING THE C2 COEFFICIENT

#### 3.1 SYLVIA modeling of aerosol deposition on walls under fire condition

The SYLVIA software, validated by the quantitative experimental data, is used to build the knowledge base of the expert system [4]. This two-zone code is designed to predict the behaviours of mechanical/natural ventilation, fire growth, hot gas and smoke propagation, and airborne contamination transfer in confined and mechanically ventilated enclosures. Within SYLVIA, each compartment is modelled by two Lagrangian control volumes separated by a thermal interface. Mass and energy balances are solved independently in each zone: the lower zone representing fresh gas, and the upper zone containing combustion products and plume-enriched gases. In a two-zone approach, a fire plume supplies the upper zone of the fire room, causing its volume to increase and thus lowering the interface. This may lead to under-oxygenation at the fire source if the exhaust duct or openings do not provide sufficient ventilation to remove the gases delivered by the plume. Ambient properties such as pressure, temperature, and species mass fractions are assumed to be uniform within each control volume.

Literature aerosol deposition models generally assume that particles are spherical or quasi-spherical. While this may approximate some aerosols, it oversimplifies the complex fractal morphology of many aerosol particles (*Figure 1 (b)*). For fractal aggregates, equivalent sphere diameters are commonly used to represent aerosol size. In addition, combustion aerosol morphology and composition vary considerably during the fire due to the fire confinement and fuel heterogeneity, which further limit the applicability of available aerosol deposition models. In this study, radionuclide release is associated with microdroplets generated during thermal degradation of TBP/HTP, which subsequently produce dry residues after passing through the flame. Ideally, input parameters include Pu mass concentration in the solution, droplet size distribution, and dry residue density which depends on the radionuclides physicochemical form. Since these parameters remain insufficiently characterized, dry residues are instead described using a log-normal size distribution defined by AMMD (*Aerodynamic Mass Median Diameter*) and geometric standard deviation, which serves as input for the expert system.

For a given aerosol population, the aerosol mass deposited on various room surfaces over time can be expressed as:

$$m_{dep} = \int_0^t C_p(t) \sum_i V_i(t) S_i dt$$

where  $C_p$  [kg/m<sup>3</sup>] is the airborne particle mass concentration,  $V_i$  [m/s] is the deposition velocity associated with mechanism  $i$  (thermophoresis, sedimentation, Brownian diffusion and inertial impaction on the ceiling in SYLVIA), and  $S_i$  [m<sup>2</sup>] is the corresponding deposition surface area.

#### 3.2 Expert system architecture

An expert system [5] has been developed to assess the fraction of radionuclides, in aerosol form, not deposited on the fire room walls (C<sub>2</sub> coefficient). The expert system is an AI tool that enhances understanding of a facility's behavior in specific situations by deriving the most likely diagnosis or prognosis in a negligible time. It combines knowledge of the underlying physical phenomena (the knowledge base) with facility-specific information. Information propagation occurs via an inference engine, i.e., the algorithmic core of the expert system based on the Bayesian Belief Network methodology. The knowledge base was built from Monte-Carlo simulations performed with SYLVIA software. The minimum size of the database depends on the number of parameters, their discretization, and the number of realizations needed to estimate the conditional probability tables. These tables encode the strength of the link between the parameters and the expert system outputs. Given the number of parameters used here (8 parameters and 5 outputs) and their discretization, a database of 7 million SYLVIA calculations was generated (requiring 2 days of CPU time spread over 1024 cores). The expert system's Graphical User Interface is shown in *Figure 5*. It displays the parameter names and value classes (top), and the output classes (bottom). For each class, a checkbox indicates whether that class is involved in the current inference, while the third column shows the

associated probability of occurrence. A button in the center launches the inference engine. In this example, the expert system is used in combined prognostic and diagnostic mode. The query submitted is: Are there any cells in the F workshop where the  $C_2$  coefficient is between 0.7 and 0.75, for a tank leak rate below 12% and a failure-free closure of the fire damper at inlet? The expert system identifies one cell (05), with a probability of 99%.

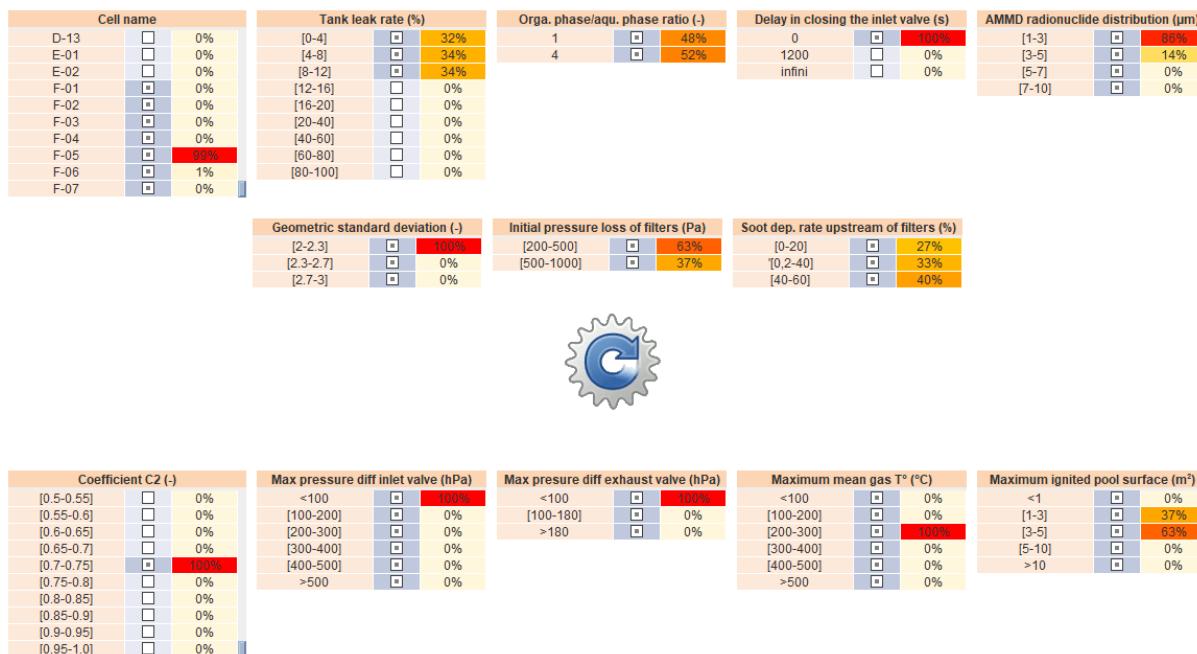


Figure 5: Graphical User Interface of the expert system

## 4 SUMMARY

In this paper, we presented recent research on the characterization and development of experimental and numerical tools to determine the  $C_2$  coefficient. First, the resistive sensor used to collect and detect aerosol particles produced by a TBP/HTP mixture was qualified at analytical scale, and a prototype was designed for implementation during large-scale fire events. Then, the new dynamic multi-sensor system, based on measuring the  $\text{CO}_2$  emitted during regeneration of the aerosol deposited on the resistive sensor, was described. Finally, an AI-based expert system was developed to assess the  $C_2$  coefficient as part of the ASNR methodology for evaluating the fire source term within a joint research project with Orano. All these tools will be tested during real-scale experiments scheduled for 2027 in PLUTON Facility.

## ACKNOWLEDGEMENTS

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