AIRBORNE RESUSPENSION OF A PLUTONIUM DIOXIDE SURROGATE DURING THERMAL DEGRADATION OF POLYCARBONATE PANELS USED ON GLOVE BOX STRUCTURES IN NUCLEAR FACILITIES

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TITLE

AIRBORNE RESUSPENSION OF A PLUTONIUM DIOXIDE SURROGATE DURING THERMAL DEGRADATION OF POLYCARBONATE PANELS USED ON GLOVE BOX STRUCTURES IN NUCLEAR FACILITIES

RESUME

Nous présentons une étude expérimentale sur la dégradation thermique de panneaux en polycarbonate (PC) représentatifs des parois de boîtes à gants rencontrées dans les usines de fabrication ou de retraitement des combustibles nucléaires. Les échantillons de PC sont contaminés par des particules de dioxyde d'hafnium (HfO_2) utilisé comme simulant non radioactif du dioxyde de plutonium (PuO_2). Les essais réalisés dans un dispositif de dégradation thermique développé à l'IRSN révèlent des fractions de remise en suspension dans l'air (ARF) de l'ordre du dixième de pour cent.

ABSTRACT

We present an experimental study on the thermal degradation of polycarbonate panels (PC) representative of the glove box walls encountered in nuclear fuel fabrication or reprocessing plants. The PC samples are contaminated with hafnium dioxide (HfO_2) particles used as a non-radioactive surrogate of plutonium dioxide (PuO_2). The tests carried out in a thermal degradation device developed at the IRSN reveal airborne resuspension fractions (ARF) of the order of a tenth of a percent.

MOTS-CLÉS : Feux de boîte à gant, resuspension, dioxyde de plutonium / **KEYWORDS:** Glove box fire, airborne resuspension, plutonium dioxide

1. INTRODUCTION

Fire is identified as the most expected damage scenario for the glove boxes (GB), in which the MOX nuclear fuel, consisting of a mixture of plutonium oxide and uranium oxide, is handled in fuel assembly and recovery plants. The walls of these GB, in part made of polycarbonate (PC), can contain labile radionuclides, in particular plutonium oxide (PuO_2). To characterize the airborne resuspension of radionuclides associated with GB fires, IRSN initiated the FIGARO (Fires Involving Glove boxes with Aerosol Release Occurences) research program, which aims to enhance the knowledge on the quantity of radionuclides in suspension during GB fires. The mitigation of the airborne suspension of radionuclides such as PuO_2 is of particular importance because of their ability to reach the human respiratory tracts.

This paper presents a small-scale study on the airborne release fraction (ARF) of PC sheets contaminated with hafnium dioxide used as plutonium dioxide surrogate in a thermal degradation device developed at IRSN.

2. MATERIAL AND METHOD

The PC sheets are contaminated using a powder dispersion device to reach particle surface density deposits representative of real GB. The thermal degradation device is equipped with a radiant panel and a ventilation system for the particles (surrogate and mostly soot) transfer to the sampling zone and to the exhaust. The walls of the apparatus downstream of the thermal degradation zone is heated at 150°C to avoid particle losses due to thermophoresis. The sampling is then done on polycarbonate membrane filters positioned after the heated zone (Figure 1).

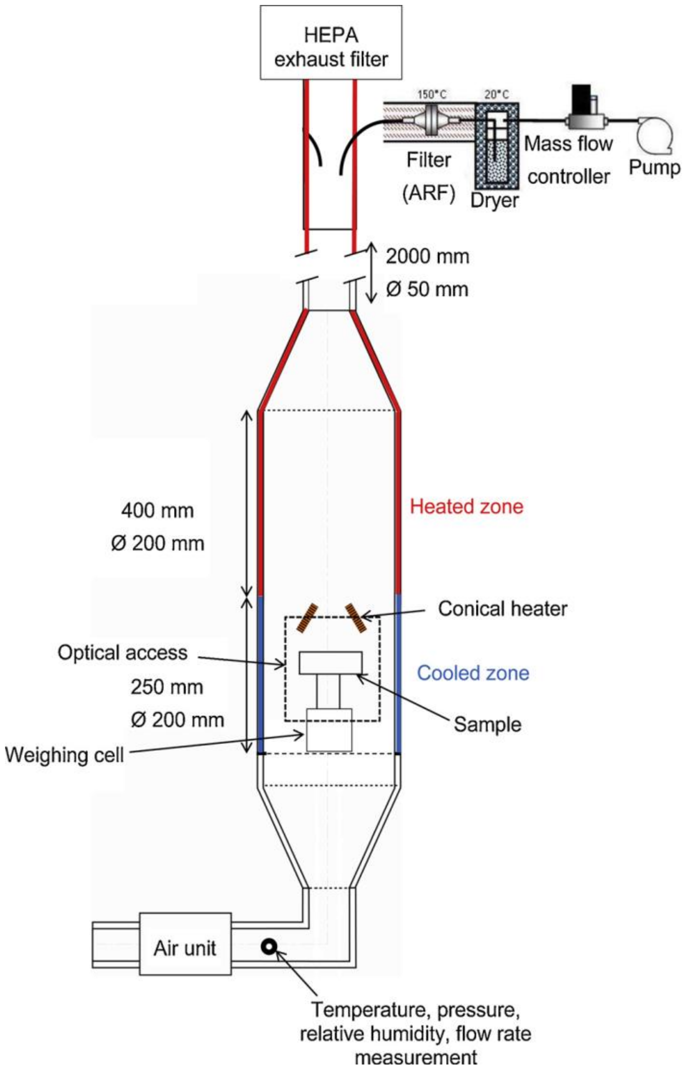

Figure 1. Schematic representation of the thermal degradation device used to assess the ARF

To carry out experiments without radioactivity, experimental investigations were done to identify the best PuO_2 surrogate. Similarity criteria based on PuO_2 density (11480 kg/m^3), as well as chemical proximity, led to the choice of hafnium oxide (HfO_2), of density 9680 kg/m^3, as the suitable surrogate. In addition, its non-radioactive nature, and its thermal stability, for fire temperatures encountered experimentally in GB at 1200 °C, substantiate the preference of HfO_2 over ruthenium oxide RuO_2, among other candidate materials. The physical characteristics of the hafnium oxide surrogate are presented in Table 1.

Table 1. Physical characteristics of the hafnium oxide surrogate

Density (kg.m^{-3})	9680
Fusion temperature (°C)	2774
Volume median diameter (VMD, coulter Coulter) (µm)	1.81
Aerodynamic median diameter (AMD, calculated from VMD Coulter) (µm)	5.63
Optical median diameter (OMV, diffractometer) (µm)	1.57

The PC sheets are contaminated by a powder dispersion apparatus to reach particle surface density deposits representative of real GB. It consists of an assemblage of a powder dispersion chamber and a cylindrical vacuum bell, in contact with the surface onto which the surrogate particles are deposited. Its operating principle is based on a sealed system, which is vacuum pumped at 100 mbar followed by a rapid air admission through a dispersion tube after the opening of a manual valve. This abrupt air admission focused toward a powder pile aerosolizes it and deposits the particles onto the polymer sheet.

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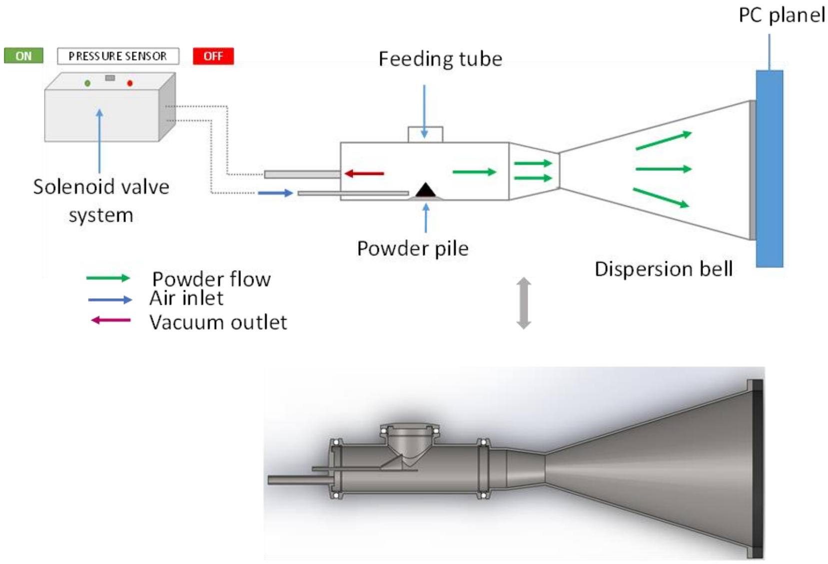

Figure 2. Schematic description of the powder dispersion apparatus

The aerosol resuspension is evaluated using the ARF which is defined as the ratio between the mass of surrogate hafnium oxide collected on the membrane filter ($m_{collected}$) as given by ICP-MS analysis and the initial deposited mass on the PC ($m_{deposited}$):

$$ARF = \frac{m_{collected}}{m_{deposited}}. \qquad (1)$$

3. RESULTS

In this study, PC sheets of 9 x 50 x 50 mm^3 were contaminated on their whole surface area of 25 cm^2. The surface density of the deposit obtained on 6 samples is on average 1.58 mg/cm^2, with a coefficient of variation as low as 0.08. This density is consistent with PuO_2 contamination levels reported in fuel reprocessing glove boxes evaluated from few $\mu m/cm^2$ to few mg/cm^2 (Ouf et al. 2020).
The contaminated PC samples are placed in the thermal degradation zone in arrangements either backside or frontside of the radiant panel (Figure 3).

Figure 3. A contaminated PC sheet in the thermal degradation zone. On the right of the figure, we can see a protective mask which isolates the sample from the heat flow before the start of the test

The radiant panel is operated at a radiation flux of 40 kW/m^2. The sample is placed at 25 mm from the radian panel and the device flowrate is 40 m^3/h along with filter sampling flowrate of 10 L/min.
During the tests, we observe a first phase associated with the formation of small bubbles on the surface of the polymer, followed by an inflammation of the sample for about 5 min. The latter ends up collapsing due to its

deformation after reaching its vitreous transition temperature. The sampling of the aerosol is carried out over a total time period of 30 min which starts as soon as the sample comes into contact with the radiant panel by removing the protective mask which isolates it from the radiative flux.
The assessed ARF values lean between 0.14 and 0.24, with no noticeable influence of the sample arranged either in the backside or in the front side of the radiant panel (Table 2).

Table 2. ARF assessed on 6 contaminated PC sheets

Config.	Trial	Mass of filter (mg)	ICP-MS (µg/filter)	ARF (%)
Backside	1	11.6	0.9	0.15
	2	15.7	1.2	0.20
	3	14.1	1.4	0.24
Frontside	1	7.4	0.8	0.14
	2	13.5	0.8	0.14
	3	8.1	0.8	0.14

4. CONCLUSIONS

Our study shows that in the event of accidental glove box fires in fuel fabrication or reprocessing plants, a subsequent airborne resuspension of plutonium oxide particles would occur. The aerosol resuspension fractions provided in this experimental work, of the order of a tenth of a percent. To go further, these values will be consolidated by large-scale tests using real GB, which will be carried out on the experimental facilities of the IRSN's GALAXIE platform.

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