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Airborne release of hazardous micron-sized metallic/metal oxide particles during thermal degradation of polycarbonate surfaces contaminated by particles: Towards a phenomenological description.

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Graphical abstract

Highlights

- Experiments concerning the airborne release of micron-sized metal particles are presented.
- Thermal degradation of particulate-contaminated polycarbonate surfaces is considered.
- The influences of the particle diameter, density and number of mono-layers are reported.
- A semi-empirical factor is proposed to consider the common influence of size and density.
- A phenomenological description of the release of particles from PC surface is proposed.

Abstract

The release of radioactive particles during fires is a key issue for the safety analysis of industrial nuclear facilities. Nevertheless, significant discrepancies exist between experimental measurements reported in the literature of airborne release fractions (ARF), expressed in terms of mass, and further discussions on the phenomenology of particles released from burning solid surfaces are needed. Experimental results are reported on the resuspension of metallic/metal oxide particles deposited on polycarbonate (PC) samples, representative of glove boxes used in the nuclear industry, under thermal degradation and for several particles deposit properties, i.e. equivalent volume diameter (D_{ev}), density (ρ_p), morphology and number of mono-layer (N_{mono}). A significant influence of D_{ev} and ρ_p was identified, with a peak in ARF for diameters close to 6 μm and a decreasing ARF with increasing density. Furthermore, the particle deposit structure was identified as an influencing parameter, with ARF decreasing with increasing N_{mono} up to nearly 0.3 and remaining constant above. Experimental results obtained in this study were compared with literature values to propose a phenomenological description of particles resuspension from burning PC surfaces. These findings open the way to a theoretical description of airborne release and to propose realistic surrogate to conduct large-scale fire experiments.

Keywords: Airborne release fraction, polycarbonate surfaces, combustion, particulate deposit.

Introduction

The recent dramatic fire of the Notre-Dame Cathedral (15-16 April, 2019 in Paris, France) has highlighted the need for a precise prediction of consequences of such fires in terms of the dispersion of potentially hazardous materials in the form of particles and, in this specific case, in the form of toxic lead particles released from the burnt spire of Notre Dame. Similar attention is paid to resuspension and the atmospheric transport of radioactive particles during wildfires in contaminated surroundings of the Chernobyl site (Evangelidou et al., 2016; Yoschenko et al., 2006). Apart from these health and environmental considerations, there is a lack of knowledge on the evaluation of the aerosol release associated with fire events involving hazardous materials within industrial facilities. Such data are primarily important in order to propose realistic contamination level and corresponding exposure doses of workers during their fire interventions or post-fire investigations in hazardous facilities (Brown, Zepper, Louie, & Restrepo, 2015; Plagge, Krause, Riva, Schäfer, & Forkel-wirth, 2017; Zepper, Brown, Pierce, Voskuilen, & Louie, 2017). Furthermore, source terms are also needed as input data for atmospheric transport simulations (Sarkar et al., 2017) to predict contaminated aerosol deposition and estimate the toxicological and ecotoxicological impact of such contamination (Bryant, 2019; Kinoshita et al., 2011).

During the production and waste treatment of nuclear fuel rods, mixed oxide (MOX) powders composed of plutonium dioxide (PuO_2) and uranium dioxide (UO_2) are handled within static containment devices such as glove boxes (Dorrian & Bailey, 1995; Massiot et al., 1998). Under normal operations, glove boxes are designed for handling radioactive material in all its forms and for providing containment of such material. Throughout their life cycles, the handling and processing of fuel rods induces the emission and release of particulate contaminants, in the form of micron-sized particles, which can be transported and deposited on glove box walls mainly composed of polymers (polymethyl methacrylate PMMA and/or polycarbonate (PC)). These materials are generally used for their good mechanical properties and their transparency to enable monitoring or carrying out operations within the glove boxes. Nevertheless, their thermal stability is significantly lower than metallic or

glass panels, which must be considered during safety analyses and especially in the event of a fire occurring inside the glove box (through pyrophoric reactions/solvents ignition) or outside the glove box from another heat source (electrical cabinet malfunctions, garbage fire etc.). Several authors have considered such hazardous situations (Buijs, Chavance de Dalmassy, & Schmidt, 1983; Fernandez & Burghoffer, 1995; Mishima & Schwendiman, 1973; Ouf et al., 2013; Quintiere, 1998; Seehars, 1983; Van Geel, Magill, & Schmidt, 1992), but few of them have discussed the physico-chemical properties (size distribution, morphology and composition) and structure of particles contamination within industrial glove boxes and more specifically of the particles physically bonded to the surface of their inner polymeric walls. Of course, since many different facilities and processes are represented in the nuclear industry, it is impossible to identify one single fire scenario involving glove boxes for which a consensus can be reached regarding contamination levels on the glove box walls and deposits on the glove box floor.

A review conducted by Dorrian & Bailey (Dorrian & Bailey, 1995) reports on an overview of 52 past publications (1967-1993) concerning the measurement of the AMAD (activity median aerodynamic diameter) of radioactive aerosols in nuclear facilities and laboratories. The paper also surveys the characterization (using a variety of analytical methods) of PuO₂ particles contained in glove boxes. The set of results examined by these authors shows that the plutonium dioxide particles found in nuclear facilities are in the form of micron-scale particles. In the case of PuO₂ particles in glove boxes, the average aerodynamic diameter of the particles is generally 4.0 μm (associated with a geometric standard deviation, σ_g , of 2.21), with the specific surface area ranging from 1 to 15 m²/g (Machuron-Mandard & Madic, 1996) and having complex morphologies (Duncan & Kane, 2009; D. Wayne et al., 2008). Such conclusions are in good agreement with the characterization of ²³⁸PuO₂ particles released from a glovebox system in the Plutonium Facility at Los Alamos National Laboratory (Cheng et al., 2004) and also with a recent analysis of plutonium oxides produced from the direct oxidation of Pu metals (Wayne, Peppers, Schwartz, & DeBurgomaster, 2018).

Another important point concerning the actual situation in Basic Nuclear Facilities (BNFs) is the surface concentration of the contaminant deposits. As is the case with physico-chemical properties of PuO₂ powders, the nature of the deposits varies greatly between different facilities. However, a literature analysis conducted at IRSN (Ouf et al., 2015) has highlighted the fact that PuO₂ deposits on glove box walls tend to be homogeneous, relatively mono-layered and with contamination levels ranging from a few $\mu\text{g}/\text{cm}^2$ to a few mg/cm^2 . In the present work, by aiming to describe particle resuspension in the event of fires involving polycarbonate panels contaminated by PuO₂ particles, we consider these orders of magnitude as relevant. For additional details, we have presented, in supplementary information Table SI_1, a non-exhaustive overview of the physico-chemical properties of PuO₂ deposits, representative of several nuclear facilities and as also reported in previous experimental studies.

Apart from the definition of the nature and contamination level, the second major point of discussion when considering radiological consequences of fires in nuclear facilities remains in the airborne release fraction (ARF) values to be considered. In general, this ARF is defined as the ratio between the mass of particles released in the airborne phase (in the form of an aerosol) and the mass of particles initially deposited on the surface under consideration. Recently, we have proposed (Ouf et al., 2013) a review of values reported in the literature for realistic materials and significant discrepancies have been discovered for fuels representative of glove boxes (i.e., polymers like PMMA or PC). Due to their high radiotoxicity (Muggenburg et al., 2008; Voelz & Buican, 2000), PuO₂ or UO₂ powders cannot be used for conducting experimental studies regarding their resuspension potential under the complex thermal stresses representatives of fires. Nevertheless, in most of the previous studies, few details have been presented regarding the criteria associated with the choice of surrogates for PuO₂ or UO₂ particles. The aim of the present paper is to describe experimentally, the phenomenology of release of particles in the event of thermal degradation of polycarbonate surfaces, one of the most representative polymers used in the nuclear industry for manufacturing glove boxes, that are contaminated by physically bonded micronic PuO₂ particles.

Regarding literature specifically available for this configuration, few data are available concerning the airborne release fraction ARF, defined as the ratio between the mass of particles released in the airborne phase (m_{released}) and the mass of particles deposited (m_{initial}) on the polymer surface:

$$\text{ARF} = \frac{m_{\text{released}}}{m_{\text{initial}}} \quad (\text{Eq. 1})$$

Initial experiments, using PC contaminated by PuO_2 particles, were conducted by Pickering et al. (1989). Mass median aerodynamic particles ranging from 1 to 9 μm were considered and cylindrical small samples (20 mm length and 84 mm diameter) were contaminated by 1 mg/cm^2 through rolling them in powder pre-classified in terms of size using a cascade impactor. The corresponding ARF decreased from $3.0 \cdot 10^{-3}$ to $4.0 \cdot 10^{-4}$ as equivalent volume diameter, D_{ev} , (computed assuming spherical particles with density of 11,460 kg/m^3) increased from 0.5 to 2.4 μm .

In parallel, the same team also conducted large scale experiments (Buijs, Chavane de Dalmassy, & Baumgärtner, 1992; Buijs, de Dalmassy, & Pickering, 1989) with PuO_2 surrogate (mixture of cerium oxide and europium oxide with D_{ev} of 3.8 μm). The ARF was reported as decreasing from 0.8 to 0.2 % as surface mass deposits increased from 0.5 to 1 mg/cm^2 . More recently, our team has also performed experiments with small scale PC samples ($10 \times 10 \text{ cm}^2$), using micronic alumina oxide particles with equivalent volume diameter, D_{ev} , of 0.9 and 3.8 μm . The surface mass concentration of the deposit was fixed at 1 mg/cm^2 and the ARF was reported to increase from 3.2 to 6.9% with increasing D_{ev} (Ouf et al., 2013).

Despite the limited number of studies addressing this complex topic, we have tried to identify several physico-chemical properties of the powder deposit which may strongly influence the release phenomena and the corresponding ARF. For a supporting discussion of experimental values reported in the literature, a phenomenological description of particle behavior at the polycarbonate surface during its thermal degradation is proposed in Figure 1. Prior to thermal degradation, the particle deposit is characterized by the size distribution, density and morphology of the particles it is composed of. Such properties also drive the structure of the cake deposit from mono to multilayered, with cohesion forces increasing with the number of layers and, as a consequence, the point of contact between particles. When applying an external heat flux to the surface, polycarbonate melts from a solid to a viscous phase, thereby trapping particles within the molten PC. In the meantime, particles still presenting at the surface of the molten PC may be released by aerodynamic forces induced by convective flow. Finally, due to its specific nature, PC forms an intumescent char at its surface, keeping the trapped particles within this porous structure and subject to complex airflows within its vicinity.

To our knowledge, there is no phenomenological model currently available of the release of particles for such situations and experimental results associated with this complex scenario are limited. Nevertheless, the available literature has assisted us in defining deposit properties for consideration within the present parametric study.

Firstly, particle geometric diameter (or equivalent volume diameter) is suspected to directly influence the interactions between particle and the PC surface during its thermal degradation. Nevertheless, the limited data reported in the literature come to controversial conclusions, with an enhancing effect with increasing volume equivalent diameter found by Ouf et al. (2013) while Pickering et al. (1989) report an inverse effect of particle diameter. The expected size effect could be associated with adhesion forces at the surface of polycarbonate sample during its thermal degradation.

Figure 1: Schematic illustration of particle behavior at a PC surface during degradation under thermal stress

In relation to any potential effect of particle density, the ARF reported for particles with equivalent volume diameter of nearly 3 μm ranged from nearly 3% for Al_2O_3 (lowest particle density 3,960 kg/m^3 from Ouf et al., 2013), 0.2% for Cerium-Europium oxide (intermediate particle density of 7,421 kg/m^3 estimated from Buijs et al., 1992a, 1989) and finally 0.0004% for 80% UO_2 – 20% PuO_2 mixed powders (highest particle density of 11,068 kg/m^3 , Pickering, 1989). This development over several decades highlights the significant contribution of particle density to the release phenomenon, mainly due to settling of those contaminant particles within the melted polycarbonate during its thermal degradation.

The aim of the present study was to perform an experimental parametric study on the influence of these deposit properties (particle diameter, density, specific surface area, morphology, and also the number of mono-layer) on the final airborne release fraction. An analysis of the results obtained and a comparison with values in the literature was then used to propose the phenomenology of release of particles from burning PC surfaces. Finally, the major influencing properties to be used when identifying a good surrogate of PuO_2 powders for conducting measurements of ARF during realistic and large scale glove boxes fires were introduced.

1. Experimental procedure

a. Particle contamination and materials

This parametric study was proposed in order to investigate the influence of powder deposit properties on the ARF. Thirteen powder samples were considered and characterized in terms of size distribution with a TSI Aerosizer PSD 3603 to measure the mass median aerodynamic diameter (MMAD), with an optical automatic microscope Malvern Morphologi G3 used to measure the projected surface area equivalent diameter (D_{psac}), in terms of morphology using scanning electron microscopy (JEOL JSM 6010-LV) and in terms of specific surface area by BET analysis (Micrometrics© ASAP 2000). The densities were considered from manufacturer data and the samples properties are summarized in Table 1. Low magnification SEM images are also available in the supplementary information SI3 for all studied samples. Zirconia oxides nanopowders were analysed using a Beckman LS13 320 laser diffraction particle sizer due to their nanometric size range under detection limit the aforementioned analyzers. The median geometric diameter D_{geo} was determined from the MMAD and particle density, and they were considered to be spherical: $D_{\text{geo}} = \text{MMAD} \cdot \sqrt{\rho_0/\rho_P}$ (with ρ_0 and ρ_P as the reference density of 1,000 kg/m^3 and the particle density, respectively).

Polycarbonate samples (PC, LEXANTM 9030-112, General Electric, 50 x 50 x 9.5 mm^3) were considered as representative polymers composing the containment barrier of glove boxes used in the nuclear industry. Sample contamination was carried out using a specifically designed device composed of a low pressure powder disperser to produce deposits with a surface mass concentration ranging from 0.01 to 1 mg/cm^2 . Small amounts of powder

(100-300 mg) were placed in a pile in a 40 mm inner diameter stainless steel tube connected to a vacuum pump, and also to a dispersion bell with an internal diameter of 12 cm. PC samples were fixed on a Plexiglas plate placed in contact with the dispersion bell and low pressure (100 mbars of absolute pressure) was applied to the system. A stainless steel tube with a 6 mm inner diameter, placed in front of the powder pile, was connected to a valve that was rapidly opened, resulting in turbulent and rapid air entry in the tube and efficient dispersion of particles on the PC samples. A determination of the mass deposit was carried out by direct mass measurement of the PC samples prior to and after particles dispersion using the contamination device. Contamination levels were then characterized by their mass to surface area concentration, defined as the ratio between the mass of particles deposited on the polycarbonate sample and the surface area of this sample (50 x 50 mm²).

An illustration of alumina particle (SPM102) deposits produced using this device is shown in Figure 2. In order to compare results obtained for powders presenting significantly different density and size distributions, we also computed, for each contaminated sample, the number of mono-layer N_{mono} of the mass of particles at the sample surface. The details regarding the computation of N_{mono} are available in the supplementary information (SI2). As reported in Figure 2, the particle deposit is composed of a mono-layer in most cases. Nevertheless, for a mass to surface area concentration higher than 0.34 mg/cm², the probability that one particle makes contact with another tends to be non-negligible.

1) 0.09 mg/cm ² / N_{mono} = 0.08	2) 0.19 mg/cm ² / N_{mono} = 0.17	2) 0.34 mg/cm ² / N_{mono} = 0.31

4) 0.42 mg/cm ² / N_{mono} = 0.38	5) 0.65 mg/cm ² / N_{mono} = 0.59	6) 0.77 mg/cm ² / N_{mono} = 0.70

Figure 2: SEM images of SPM102 alumina particles deposits on PC samples

b. Contact angle measurement setup

As previously mentioned, interactions between particles and the polycarbonate surface during its thermal degradation play major role in the stability of such particles at the air-particle-PC interface and, as a consequence, on their availability for any further airborne release due to external forces. Apart from this simplified description of particle fate at the surface of a polymer subjected to heat flux exposure, measurement of the contact angle produced between molten polycarbonate and a particle deposit could be helpful for any future phenomenological development. To determine this contact angle by experiment, assistance was sought from the company KRÜSS®, an expert in contact angle measurement. The experimental apparatus (KRÜSS® Drop Shape Analysis System DSA100¹) relies on the analysis of images obtained using the sessile drop technique. A temperature controlled chamber (TC21) was also used to take measurements at a higher temperature (200-210°C) than the glass transition temperature of polycarbonate (150°C). Details on measurement protocol are available in the supplementary information SI4. Alumina oxide, tungsten and tungsten

¹ <https://www.kruss-scientific.com/products/contact-angle/dsa100/drop-shape-analyzer-dsa100/>

oxide particles were considered, as they present projected surface areas equivalent diameters of 10 μm , 8 μm and 3 μm with angular, spherical and cluster shapes respectively. The characteristics of the deposits studied are explained in detail in Table SI4-1. For each sample, 6 measurements of the angle of contact between particles in a mono-layer deposit (on a glass slide) and one drop of molten polycarbonate were taken. The number of mono-layer was determined from the mass deposited per unit area (the deposit was assumed to be homogeneous on the surface of the glass slide measuring 19.76 cm^2) using the method explained in SI2.

c. Combustion test bench

Thermal degradation and ARF measurements were carried out on an experimental test bench called ARTEMIS (a French acronym of “Appareil de dégradation Thermique pour l’Etude de la Mise en Suspension” and translated into English by “Apparatus of thermal degradation for studying airborne release”), specifically designed by our team for limiting losses of released particles during their transport from the emission to the sampling point. Figure 3 summarizes the main principle of the thermal degradation and sampling devices. A ventilation fan coupled to a critical orifice allows the injection of air at a fixed flow-rate (from 40 to 60 m^3/h) inside the degradation chamber composed by a radiant conical heater. Particles were then extracted from this chamber inside an exhaust duct heated at 150°C, before being sampled under well-mixed conditions at 2 meters from the inlet. Finally, exhaust gases and particles were filtered with an HEPA filter before releasing it into the atmosphere. In previous studies, most of the devices were not optimized to reduce losses by deposition on the walls; this may explain the difficulties encountered in obtaining a proper mass balance and the disparities in terms of ARF between different studies even for the same fuel. As a consequence, our test bench has been optimized for reducing losses of particles in the combustion chamber and the exhaust duct. In addition, transport efficiency measurements and CFD computations were carried out for the same particles as those deposited on the polymers samples and the ARF values were corrected according to these losses (see SI5). The particles released were sampled on cellulose membranes with an isokinetic probe and the ARF were determined by ICP-MS/AES. Using this test bench, experiments were conducted for particles of different sizes, natures, densities and specific surface areas for radiative heat fluxes ranging from 15 to 50 kW/m^2 . In the present case, the heat flux was fixed to 50 kW/m^2 to promote the auto-ignition and burning of PC samples.

Table 1: Physico-chemical properties of powders considered as contaminant

Nature	Name	MMAD (μm) σ_g (-)	D_{geo} (μm)	Density (kg/m^3)	S_{BET} (m^2/g)	Morphology	SEM image
Alumina Al_2O_3	SPM84	25.9 / 1.8	13.0	3,950	-	Angular, Facets
	SPM102	6.4 / 2.2	3.2		1.7		
	SPM105	7.0 / 2.1	3.5		-		
Cerium oxide	US NANO	15.3 / 2.5	5.5	7,650	-	Long, thin needles

CeO ₂	AAT	21.2 / 2.6	7.7			Angular, bulky
Tungsten carbide WC	WC1	1.4 / 1.7	1.7	15,630	1.3	Facets, low porosity
Tungsten oxide WO ₃	US NANO D4	10.5 / 2.2	3.9	7,160	-	Submicron aggregates
	D17	23.4 / 1.7	8.7			Angular, bulky
	D27	31.2 / 1.9	11.7			Smooth angle, bulky
Ytterbium oxide Yb ₂ O ₃	US NANO	7.8 / 2.3	2.6	9,170	-	Facets, angular
Zirconium oxide	CZE-S1	-	0.20	5,680	6.1	Submicron aggregates (nanopowder)
	CZE-M1		0.24		14.7		
ZrO ₂	CZE-G1		0.20		26.5

Figure 3: Principle and illustration of the combustion test bench

A small part (reported as $P_p(\%)$ in this study and defined as the ratio between volumetric sampling flow rate and exhaust flow rate) of the overall exhaust flow was sampled and the transported particles were then deposited on a high efficiency particulate air membrane (Cellulose acetate, SARTORIS 11106-47-N, 47 mm diameter with 0.45 μm pore diameter). The mass of released particles was then determined through ICP-MS/AES analysis to compute the airborne release fraction for each experimental condition.

Despite the specific design of this test bench for reducing, to the greatest possible extent, the particle deposit within the exhaust and sampling, we were not able to consider losses of such particles as negligible and the final definition of the airborne release is:

$$ARF = \frac{m_{\text{particles}}^{\text{sample}}}{m_{\text{initial}} \cdot P_p(\%) \cdot F_p(\%)}, \quad (\text{Eq. 2})$$

with:

- $m_{\text{particles}}^{\text{sampling}}$ as the mass of released particles determined by ICP-MS/AES on the sampling membrane,
- $F_p(\%)$ as the penetration fraction of particles within the ARTEMIS device dependent on the aerodynamic diameter of released particles.

The mass of released particles was then defined as a function of the mass of released particles determined on the sampling membrane, corrected for losses of particles within the test bench according to the penetration factor $F_p(\%)$. It was then crucial to determine this penetration factor for each type of particles deposit. Two different approaches were considered for this purpose: one experimental and one based on CFD computations. More details on these mutual approaches are available in the supplementary information (SI5). For the first one, powder samples used for particles contamination were injected in the ARTEMIS test bench, using a rotating

brush powder disperser (RBP 1000 PALAS[®]), above the polymer sample. The penetration factor was determined by filter sampling, directly at the outlet of the injection point and at the sampling point. The penetration factor was defined as the ratio between the mass flow rate of particles at the sampling point divided by the mass flow rate of particles at the injection point.

On the other hand, CFD computations were carried out for similar experimental conditions with ANSYS CFX and particle transport models (Nerisson et al., 2011), and recently implemented by Gelain et al. (2018). Measurements and computations were conducted for all conditions considered (see figure SI5-3) and corresponding penetration fractions were considered for the final determination of ARF.

d. Experimental protocol and analysis of samples

The protocol for the resuspension experiments was based on several steps. Polymer samples with particle deposits were placed in the combustion chamber and the test started when the heat flux was applied at time (t_0). Aerosols were sampled on filters sequentially during experiments. The polymer sample residue was then weighed and analysed in terms of particle trapping (see SI) and aerosol filters were analysed by ICP-MS after each test.

Special care was given to the ICP-MS analysis protocol and particularly to the mineralization step. For most samples, acid digestion was considered for completing the dissolution of samples filters in an efficient way within a liquid medium suitable for ICP-MS analysis. For calibrating the overall ICP-MS procedure, reference filters containing a known amount of each considered powder were prepared and analysed. For this purpose, the different powder samples considered in this study were aerosolized using a vortex shaker unit and the deposited mass was determined by weighing of the membrane before and after powder dispersion. The mineralization yield of ICP-MS analysis, mainly associated to the process of dissolution or mineralization of the sample from solid to liquid phase, was then determined by comparing the mass of the considered element obtained by ICP-MS with the mass deposited on the reference membrane. According to this protocol, the mass of particles composed by the element X and sampled on membranes during experiment was defined by:

$$m_X^{\text{sample}} = \frac{m_X^{\text{ICP-MS}}}{R_{\text{detection ICP-MS}}^{\text{chemical form of X}} \cdot R_{\text{mineralization}}^{\text{chemical form of X}}} \quad (\text{Eq. 3})$$

with:

- m_X^{sample} as the mass of element X sampled on membrane (kg),
- $m_X^{\text{ICP-MS}}$ as the mass of element X quantified by ICP-MS analysis (kg),
- $R_{\text{detection ICP-MS}}^{\text{chemical form of X}}$ as the ICP-MS yield of detection of element X (considered here to be 1),
- $R_{\text{mineralization}}^{\text{chemical form of X}}$ as the yield of mineralization of element X in its chemical form as sampled on membrane.

2. Experimental results

A set of 12 types of powder was used to carry out the parametric study (the tungsten carbide WC shown in Table 1 was not used for the PC, but is shown for comparison with PMMA, see Figure 4). For each of these samples, 5 repetitions of the resuspension experiment were carried out.

a. Influence of the number of mono-layer

As explained in the phenomenological analysis of resuspension, the surface concentration of the contaminant, or more specifically the number of mono-layer of particles present on the surface, is therefore potentially a parameter of prime importance. When increasing the number of mono-layer, one could then expect a significant decrease of airborne release fraction due to the enhancement of cohesion forces within the particle deposit. However, although this may be the case, this parameter could not be considered as a physico-chemical criterion

to be taken into account for mimicking the behaviour of PuO_2 particles since number of mono-layer composing the deposit is related to particle-particle interaction rather than particle-surface interactions, which are more relevant and of prime influence for the present investigation.

Figure 4: Normalized ARF as a function of the number of mono-layer

To compare results obtained for different powders, associated to different ARFs, the airborne release fraction of each sample was normalized according to ARF reported for the number of mono-layer close to 1, making its asymptotic value tending towards 1. Figure 4 shows this comparison, highlighting a significant decrease of dimensionless ARF as the number of mono-layer increases up to a threshold 0.31. This threshold effect obviously poses an issue when it comes to comparing the influence of properties specific to the different samples used in this study. We will therefore ignore any airborne release fraction values obtained for samples with fewer than 0.31 mono-layer in the rest of this study. Therefore, SPM84 powder will not be considered in the remainder of the parametric study because its number of mono-layer was always below this threshold value of 0.31.

Figure 4 shows the change in the airborne release fraction according to the number of mono-layer for all experiments (apart from zirconia ZrO_2 for which the analyses could not establish the quantity of zirconium above the ICP-MS detection limit).

For alumina (Al_2O_3), a significant change in the airborne release fraction can be seen for numbers of mono-layer below a threshold value of around 0.4. This change is also visible, to a lesser extent, for the CeO_2 and WO_3 samples and for deposits with a 0.3-0.4 mono-layer. A similar change can also be observed in the case of PMMA, which highlights the importance of this parameter in relation to the nature of the deposit. This threshold concept also accords well with the images of the deposits, which highlighted the significantly more cohesive appearance of deposits with more than a 0.31 mono-layer (see Figure 2). Considering these experimental changes and the appearance of the deposits shown in Figure 2, we will therefore set the threshold value in terms of number of mono-layer at 0.31.

b. ARF mean values

Taking the airborne release fractions associated only with samples with mono-layer greater than 0.31, we can determine mean values of these fractions. Looking at Figure 4, it appears that above this threshold the airborne release fraction for a single type of powder does not change significantly. The mean airborne release fraction

values associated with each sample and their corresponding combined uncertainties are given in Table 2. The highest ARF values are reported for alumina and cerium oxides. We were not able to determine any ARF for ZrO_2 since zirconia masses present on samples were lower than the detection limit of ICP-MS/AES. Apart from experimental uncertainties associated to each studied condition, several orders of magnitude were noticed between the ARFs reported. Further discussion, from a phenomenological point of view, is needed and will highlight the most influencing parameters of particles composing the deposit.

Table 2: Mean ARF values (mono-layer greater than 0.31, except SPM84)

Contaminant	D_{geo} (μm)	ρ_p (kg/m^3)	ARF (%)	2.u(ARF) (%) (95% C.I., $k=2$)
Al_2O_3 SPM84	13.0	3,950	71.23	313.49
Al_2O_3 SPM102	3.2	3,950	30.39	83.01
Al_2O_3 SPM105	3.5	3,950	72.74	23.46
CeO_2 A.A.T.	7.7	7,650	61.75	66.47
CeO_2 US NANO	5.5	7,650	85.66	144.03
WO_3 US NANO	3.9	7,160	1.12	3.24
WO_3 D17	8.7	7,160	3.61	11.27
WO_3 D27	11.7	7,160	0.31	0.55
Yb_2O_3 US NANO	2.6	9,170	3.46	3.56
ZrO_2 CZE-S1	0.20	5,680	-	-
ZrO_2 CZE-M1	0.24	5,680	-	-
ZrO_2 CZE-G1	0.20	5,680	-	-

c. Effect of particle diameter

Figure 5 shows the change in the airborne release fraction, for samples with more than a 0.31 mono-layer, according to the median geometric diameter (D_{geo}) of the particles. For the sake of a comparison, we have added this figure the results reported for cerium/europium oxide $\text{Ce}_{0.84}\text{Eu}_{0.16}\text{Ox}$ particles (Buijs et al., 1992), MOX ($\text{U}_{0.8}\text{P}_{0.2}\text{O}_2$) particles (Buijs et al., 1989), MOX ($\text{U}_{0.8}\text{P}_{0.2}\text{O}_2$) particles classed by size using an impactor (Pickering, 1989) and alumina SPM102 particles (Ouf et al., 2013) deposited on PC. The very strong influence of particle diameter is revealed for this experimental configuration (vertical orientation of contaminated samples in regard to the heat flux source, development of intumescence, and number of mono-layer greater than 0.31).

Figure 5 : The ARF as a function of median geometric diameter ((1) : Buijs et al., 1989 ; (2) : Buijs et al., 1992 ; (3) : Pickering, 1989 ; (4) : Ouf et al., 2013)

d. Measurement of contact angle

In order to investigate the influence of particle diameter on ARF, contact angle measurements were additionally conducted. From the action of a heat flux, the surface of the contaminated PC liquefies and a capillary action occurs between the particles and the polymer. In such situation, the particle diameter and morphology could influence suspension mechanisms, particularly because of their effect on the surface tension between the molten PC and the particle deposit. Because this surface tension depends on the angle of contact between these two phases, any alteration of the nature of the molten PC/particle deposit interface (number of mono-layer, particle size and morphology) would have a direct impact on this contact angle. More precisely, an increase of contact angle will reduce the wettability of the particles, reducing particle trapping and, conversely, increasing ARF.

Figure 6 shows the change in the contact angle as the median equivalent projected area diameter of the particle changes. Logically, the spherical particles (which have fewer points of contact with the molten PC) seem to have a smaller contact angle with the liquid polymer. However, it can be seen that the differences observed between the spherical particles and the angular particles or clusters appear relatively small and that morphology therefore seems to be of secondary importance compared to particle size. Concerning this second point, the results previously obtained for spherical nanoparticles of SiO₂ (McBride & Law, 2012) are shown in Figure 6. Using all these results, we also propose an empirical adjustment, which highlights the effect of the threshold associated with the particle diameter, which was suspected by McBride & Law (2012). When the diameter is lower than 1 μm, the contact angle diminishes significantly, highlighting the greater wettability of ultrafine particles and therefore their lower propensity for suspension previously reported in Figure 5. Controversially, Figure 6 does not show any significant decrease of contact angle for particles larger than 5 μm (as shown in Figure 5 for the ARF) and additional effects must be considered for explaining this peak shape of ARF vs diameter.

Figure 6 : The contact angle as a function of median equivalent projected surface diameter of particles (inset illustration from Maestro, Guzmán, Ortega, & Rubio, 2014)

e. Influence of specific surface area

Regarding the effect of specific surface area, this parameter was studied in experiments conducted with ZrO_2 nanopowders. However, considering the results obtained, we were not able to determine the airborne release fraction associated with these samples, because all the zirconia masses in the samples on the cellulose acetate membrane were below the quantification limit for ICP-MS analysis ($5 \mu\text{g}$). Due to this fact, we cannot draw any conclusions on the importance of this parameter. However, if we consider the size of the particles in the ZrO_2 nanopowders, the very limited suspension of these particles agrees with the contact angle measurements obtained by McBride & Law (2012), for spherical nanoparticles of SiO_2 deposited on polystyrene (see Figure 6). Here, the lowest values were observed for particles with geometric diameters of less than $1 \mu\text{m}$.

f. Effect of particle density

Figure 7 shows the change in the airborne release fraction depending on the density of the particles that make up the contaminant deposit on the PC surface. As shown in Figure 5, the geometric diameter of the particles has a significant influence, and a direct comparison of the results obtained for all the samples (with very different D_{geo} values) according to their density is therefore not relevant. We have therefore chosen instead to look at the configurations for which the particles have very similar geometric diameters (between 3 and $4 \mu\text{m}$). These samples, shown in red in Figure 7, underline the important effect of this parameter on the airborne release fraction and at the same time explain the dispersion reported in Figure 5 for particles with median geometric diameter D_{geo} between 3 and $4 \mu\text{m}$. A decrease of more than an order of magnitude is highlighted in Figure 7 when increasing density from nearly $4,000 \text{ kg/m}^3$ to values close to $10,000 \text{ kg/m}^3$. Such influence partially explains the decrease of ARF reported in Figure 5 for particle diameter larger than $5 \mu\text{m}$. For such high density particles, settling within melted polymers will counteract the force applied on particles by convective airflows.

Figure 7: The ARF as a function of particle density ((1): Buijs et al., 1989; (2): Buijs et al., 1992; (3): Pickering, 1989; (4): Ouf et al. (2013))

3. Discussion: toward a phenomenological description

Based on an analysis of all the results obtained in the present study and from the available literature, the airborne release fraction therefore seems to be at its maximum for a median geometric diameter between 6 and 8 μm . The airborne release fraction of particles of smaller diameters falls sharply, reaching values well below 0.01% for particles diameters lower than 2 μm . It was not possible to determine the airborne release fraction in experiments done with zirconia powders with geometric diameters of less than 0.5 μm . This result accords with the contact angle measurements presented in Figure 6, in which a significant reduction in contact angle was observed for equivalent projected area diameters (similar to geometric diameter) below 1 μm . For these particles, because the contact angle is relatively small (less than 60°), the particles are trapped in the molten polymer and the probabilities of these particles being on the surface of the molten PC and potentially released are therefore relatively small.

As the geometric diameter of the particles increases, the contact angle tends to an asymptotic value corresponding to equilibrium between the particles at the molten PC/particles/air interface (see Figure 6). Meanwhile, an increase in particle diameter causes a significant increase of their volume and, as a consequence, of the buoyant force applied on it. Therefore, for particles with a diameter of more than 1 μm , the probability of a particle being at equilibrium at the interface is non-negligible and increases with increasing diameter. Also, aerodynamic forces also increase with increasing diameter, enabling the airborne release of particles from the polymer surface. Such balance of forces could explain the very significant change in airborne release fraction observed between 1 and 6 μm in Figure 5.

A plateau is then observed just before a drastic reduction in the airborne release fraction above a threshold value of 8 μm . This reduction can be explained by the gradual trapping of the particles because they settle in the molten polycarbonate.

According to this analysis, a phenomenological description of the behavior of a particle at the air/melted polycarbonate interface was proposed in Figure 8. Considering a spherical particle with diameter D_p and density

ρ_p , capillary force and gravity force will form a meniscus around the immersed volume of the particle, which could be described by contact angle θ_c and filling angle θ_f .

Figure 8: Phenomenological description of interaction forces between particles and melted PC

Firstly, the several attractive forces are presented, defined as forces opposed to release of particles from the surface, oriented downward and including capillary F_C , gravity F_G and hydrostatic pressure F_P forces. All these forces have been previously introduced by several authors to describe the behavior of particles within colloidal systems (Shang, Flury, & Deng, 2009), particle transport at liquid interface (Joseph, Wang, Bai, Yang, & Hu, 2003) or particle-stabilised foams (Bournival, Ata, & Wanless, 2015). A simple description of those forces from an ideal situation is presented hereafter, assuming spherical particles in contact with melted polycarbonate as described in Figure 8.

Capillary force F_C is defined as a function of the radius of particle R_p forming a contact angle θ_c and filling angle θ_f with the melted PC phase denoting viscosity γ_{PC} :

$$F_C = 2\pi\gamma_{PC}R_p\sin\theta_f\sin(\theta_f+\theta_c). \quad (\text{Eq. 4})$$

Gravity force F_G is defined as a function of gravity constant g , density ρ_p and radius R_p of particle:

$$F_G = \frac{4}{3}\pi\rho_p g R_p^3. \quad (\text{Eq. 5})$$

Hydrostatic pressure F_P is defined as a function of particle radius R_p , filling angle θ_f , melted PC density $\rho_{\text{melted PC}}$, gravity constant g and immersion height z_0 :

$$F_P = \pi(R_p\sin\theta_f)^2 \rho_{\text{melted PC}} g z_0. \quad (\text{Eq. 6})$$

In addition to those attractive forces, the major repulsive (upward) force applied to particle is the buoyant force. Such force counteracts the previously introduced attractive forces and enhances the stability of particle at the interface between air and melted polycarbonate. Buoyant force is induced by liquid volume displacement associated to the immersion of particles within melted PC. The buoyant force F_B is defined as a function of particle radius R_p , filling angle θ_f and melted PC density $\rho_{\text{melted PC}}$:

$$F_B = -\frac{\pi}{3}R_p^3(1-\cos\theta_f)^2(2+\cos\theta_f)\rho_{\text{melted PC}}g \quad (\text{Eq. 7})$$

Finally, the last two forces applied to particles, F_L and F_D , lift and drag forces respectively, are introduced. Such forces have been largely described in the past by several authors to describe the aerodynamic release of micronic particles from the solid surface to the airborne phase. These two forces are defined as a function of particle radius and air density and describe the force exerted by air flow on particles when they are immersed in the boundary layer close to a solid, or in the present case, of a liquid surface.

From Cleaver & Yates (1973), Saffman (1965) and Reeks & Hall (2001), the lift force F_L is defined as:

$$F_L = 10.1\rho_{\text{air}}\gamma_{\text{air}}\left(\frac{\tau_0}{\rho_{\text{air}}\gamma_{\text{air}}}\frac{2R_p}{\gamma_{\text{air}}}\right)^3, \quad (\text{Eq. 8})$$

with γ_{air} the viscosity of air and τ_0 the shear stress at the melted PC surface.

From O'Neill (1968) and Reeks & Hall (2001), the drag force F_D is defined as:

$$F_D = 8\rho_{\text{air}}\gamma_{\text{air}}^2 \left(\sqrt{\frac{\tau_0}{\rho_{\text{air}}}} \frac{2R_p}{\gamma_{\text{air}}} \right)^2. \quad (\text{Eq. 9})$$

A theoretical description of all forces applied to particles has been introduced, nevertheless most of them need a perfect description of contact/filling angles between particles and melted polycarbonate density/viscosity (F_C , F_P , F_B), and the airflow properties within the boundary layer close to the PC surface (F_L , F_D). While such information could be retrieved for perfectly flat and non-reactive surfaces or liquids, they are not easily reachable for the present case of thermal degradation of polycarbonate. Thus, we were not able to perform computation of all those forces and we have decided to introduce a non-dimensional ratio to describe the release phenomenon of particles from our polycarbonate samples. A similar analysis has been previously proposed by Ono et al. (2017), who propose describing the suspension of particles deposited on PC using the relationship between the particle mass m_{particle} and the particle surface area S_{particle} . In the present study, and considering the proposed phenomenological description, airborne release phenomenon can be described according to the balance between repulsive (F_B , F_L and F_D) and attractive forces (F_C , F_G , F_P). Nevertheless and as previously reported, thermo-physical properties of melted PC and filling angle of particles within this complex viscous phase could not be determined experimentally. Without any further investigation, we have introduced an empirical ratio F , mainly depending on particle radius and density which are well characterized in the present study and are of main influence on ARF (see Figures 5 and 7). Since such a ratio must be relevant for representing the balance between repulsive forces (mainly ruled by particle size as shown in the left part of Figure 5 and according to equations 7 to 9) and attractive forces which are, as reported in Figure 7, directly proportional to density (eq. 5), the following form for the F ratio is proposed:

$$F = \frac{R_p^2/R_0^2}{\rho_P/\rho_0} = \frac{D_{geo}^2/D_0^2}{4.\rho_P/\rho_0} \quad (\text{Eq. 10})$$

It must be noted that, for simplification, the particle density has been divided by the reference density $\rho_0 = 1000 \text{ kg/m}^3$ and for imposing a maximum ARF for a dimensionless F ratio of 1, the particle size has been introduced in this ratio with a square power law and divided by unit diameter of $1 \text{ } \mu\text{m}$ D_0 .

Figure 9 shows the change in the airborne release fraction as a function of this F ratio and for deposits with numbers of mono-layer greater than 0.31. For particles with smaller geometric diameters (less than $2 \text{ } \mu\text{m}$ and with an F ratio below 0.1), trapping linked to the high 'wettability' of the particles seems to prevent the suspension of these particles permanently. Maximum suspension is reached at an F ratio value of 1. In this situation, the particle is stabilized at the melted PC surface by buoyant forces and therefore is available for resuspension by lift and drag forces. Beyond this threshold, settling of the particles in the polycarbonate causes them to be trapped, reducing the aerodynamic suspension.

Figure 9: Airborne release fraction as a function of F ratio ((1): Buijs et al., 1989; (2): Buijs et al., 1992; (3): Pickering, 1989))

To illustrate the phenomenology of suspension and the behavior of particles at the surface of the molten PC, the following four suspension regimes are introduced (also represented in Figure 9):

- The first regime, called the ‘contact regime’, is associated with particles with a median geometric diameter of less than 2 μm (corresponding to F ratios of less than 0.1 depending on their density), in which the particles are trapped in the molten polycarbonate and very few can therefore be released. In this case, particle stability at the melted PC/air interface is ruled by attractive forces such as capillary forces and is poorly influenced by particle size and density.
- A second regime, called the ‘aeraulic regime’, is associated with particles with a median geometric diameter between 2 μm and 6 μm (corresponding to F ratios of between 0.1 and 1.0 depending on their density). In this regime, the particles are on the surface of the molten polycarbonate and can be released mainly due to the effect of aerodynamic forces (consisting of drag and lift forces; in this case, when the polycarbonate plates are oriented vertically, the drag force is undoubtedly the dominant force). A significant increase of ARF in this range is then due to the intensity of these forces which is proportional to the median geometric diameter of the particles raised to a power (2 and 3 respectively in the case of drag and lift forces, eq. 8 and 9). This dependence thus explains the sudden increase (see Figure 5) in the airborne release fraction between 2 and 6 μm (according to a diameter power law), as well as the relevance of geometric diameter for describing this phenomenon.
- A third regime, called the ‘buoyant regime’, is associated with particles with a median geometric diameter between 6 μm and 8 μm (corresponding to F ratios of between 1.0 and 2.0 depending on their density). In such situations, buoyancy is maximal, and counteracts gravity forces leading to stability of particles at the interface enabling their aeraulic release from the melted PC surface as a consequence.
- A fourth regime, called the ‘settling regime’, is associated with particles with a median geometric diameter of more than 8 μm (corresponding to F ratios above 2.0 depending on their density), in which the particles are trapped through settlement in the molten polycarbonate.

Conclusions

The aim of this study was to identify major influencing properties of particle deposits on their airborne release fraction during thermal degradation of polycarbonate samples, which were representative of glove box panels. The experiments carried out have highlighted the significant influence of median geometric diameter and density of particles, as well as the deposit structure (mainly associated with the number of mono-layer).

In terms of phenomenology, a description was proposed that uses four suspension regimes based on the balance between repulsive and attractive forces F proportional to geometric diameter and particle density. In increasing the order of the F ratio, we introduced a contact regime associated with the trapping of particles in the molten polycarbonate, an aerodynamic regime associated with a significant increase in the airborne release fraction, a buoyant regime and a settling regime characterized by a new phase of particles trapped in the molten PC (this time through settlement).

Apart from identifying such influencing properties, the present study assists in determining a surrogate of PuO_2 in order to obtain a realistic experimental determination of ARF. For this purpose, a “good surrogate” of PuO_2 powder needs to present similar geometric diameter and density.

Investigations are currently underway to identify a surrogate of PuO_2 powder with similar properties in terms of size and density, but also on how to master the size dispersion (associated to geometric standard deviation) and morphology (close to spherical) for these reference powders. After this first step, experiments with this ideal surrogate will be conducted at an analytical scale to confirm the experimental conditions associated to the highest ARF (number of mono-layer N_{mono} lower than 0.31) and lowest ARF (N_{mono} greater than 0.31). Finally, this ideal surrogate will be considered in future research to perform large scale glove box fire experiments for the determination of ARF under real conditions.

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References

- Bournival, G., Ata, S., & Wanless, E. J. (2015). The roles of particles in multiphase processes: Particles on bubble surfaces. *Advances in Colloid and Interface Science*, 225, 114–133. <http://doi.org/10.1016/j.cis.2015.08.008>
- Brown, A. L., Zepper, E., Louie, D., & Restrepo, L. (2015). Contaminant Entrainment from a Gasoline Pool Fire. In *Fall 2015 Western States Section of the Combustion Institute* (pp. 1–20).
- Bryant, P. A. (2019). *Airborne Radioactive Discharges and Human Health Effects: An introduction*. (P. A. Bryant, Ed.) (IOP Publis). IOP Publishing. <http://doi.org/10.1088/2053-2563/aafa6d>
- Buijs, K., Chavance de Dalmassy, B., & Schmidt, H. E. (1983). Characterization of radioactive aerosols from fires. In *CSNI specialist meeting on interaction of fire and explosion with ventilation systems in nuclear facilities. Volume I* (pp. 109–117). Retrieved from http://inis.iaea.org/Search/search.aspx?orig_q=RN:15064086
- Buijs, K., Chavane de Dalmassy, B., & Baumgärtner, E. (1992). *Contaminated smoke: a simulation of the heavy metal containing aerosols from fires in plutonium gloves boxes part II*.
- Buijs, K., de Dalmassy, B. C., & Pickering, S. (1989). The dispersion of radioactive aerosols in fires. *Journal of Nuclear Materials*, 166(1–2), 199–207. [http://doi.org/10.1016/0022-3115\(89\)90190-6](http://doi.org/10.1016/0022-3115(89)90190-6)
- Cheng, Y. S., Guilmette, R. A., Zhou, Y., Gao, J., Labone, T., Whicker, J. J., & Hoover, M. D. (2004). Characterization of plutonium aerosol collected during an accident. *Health Physics*, 87(6), 596–605. <http://doi.org/10.1097/01.HP.0000138577.21388.a7>
- Cleaver, J. ., & Yates, B. (1973). Mechanism of detachment of colloidal particles from a flat substrate in a turbulent flow. *Journal of Colloid and Interface Science*, 44(3), 464–474. [http://doi.org/10.1016/0021-9797\(73\)90323-8](http://doi.org/10.1016/0021-9797(73)90323-8)
- Dorrian, M.-D., & Bailey, M. R. (1995). Particle Size Distributions of Radioactive Aerosols Measured in Workplaces. *Radiat Prot Dosimetry*, 60(2), 119–133. Retrieved from <http://rpd.oxfordjournals.org/content/60/2/119>
- Duncan, A. J., & Kane, M. C. (2009). *Properties and Behavior of 238 Pu Relevant to Decontamination of Building 235-F, Savannah River Nuclear Solutions report SRNL-STI-2009-00239*. Retrieved from <http://sti.srs.gov/fulltext/SRNL-STI-2009-00239.pdf>
- Evangelidou, N., Zibtsev, S., Myroniuk, V., Zhurba, M., Hamburger, T., Stohl, A., ... Kireev, S. I. (2016). Resuspension and atmospheric transport of radionuclides due to wildfires near the Chernobyl Nuclear Power Plant in 2015: An impact assessment. *Scientific Reports*, 6(1), 26062. <http://doi.org/10.1038/srep26062>
- Fernandez, Y., & Burghoffer, P. (1995). Radioactive Aerosols Emission in Fires. *Aerosol Science and Technology*, 23(2), 231–238. <http://doi.org/10.1080/02786829508965306>
- Gelain, T., Porcheron, E., Chagnot, C., & Roulet, D. (2018). CFD Simulations of Aerosol Dispersion and Agglomeration During the Laser Cutting of Fukushima Fuel Debris Simulants. In *Volume 8: Computational Fluid Dynamics (CFD); Nuclear Education and Public Acceptance* (p. V008T09A045). ASME. <http://doi.org/10.1115/ICONE26-82408>
- Joseph, D. D., Wang, J., Bai, R., Yang, B. H., & Hu, H. H. (2003). Particle motion in a liquid film rimming the inside of a partially filled rotating cylinder. *Journal of Fluid Mechanics*, 496(496), 139–163. <http://doi.org/10.1017/S0022112003006451>
- Kinoshita, N., Sueki, K., Sasa, K., Kitagawa, J. I., Ikarashi, S., Nishimura, T., ... Yamagata, T. (2011). Assessment of individual radionuclide distributions from the Fukushima nuclear accident covering central-east Japan. *Proceedings of the National Academy of Sciences of the United States of America*, 108(49), 19526–19529. <http://doi.org/10.1073/pnas.1111724108>
- Machuron-Mandard, X., & Madic, C. (1996). Plutonium dioxide particle properties as a function of calcination temperature. *Journal of Alloys and Compounds*, 235(2), 216–224. [http://doi.org/10.1016/0925-8388\(95\)02122-1](http://doi.org/10.1016/0925-8388(95)02122-1)
- Massiot, P., Leprince, B., Lizon, C., Le Foll, L., L'Huillier, I., Rateau, G., & Fritsch, P. (1998). Physico-chemical characterisation of inhalable MOX particles according to the industrial process. *Radiation Protection Dosimetry*, 79(1–4), 43–47. <http://doi.org/10.1093/oxfordjournals.rpd.a032444>
- Mishima, J., & Schwendiman, L. C. (1973). *Interim report: the fractional airborne release of dissolved radioactive materials during the combustion of 30 percent normal tributyl phosphate in a kerosene-type diluent*. Richland, WA (United States). Retrieved from <http://www.osti.gov/servlets/purl/4296864/>
- Muggenburg, B. A., Guilmette, R. A., Hahn, F. F., Diel, J. H., Mauderly, J. L., Seilkop, S. K., & Boecker, B. B. (2008). Radiotoxicity of Inhaled 239 PuO₂ in Dogs. *Radiation Research*, 170(6), 736–757. <http://doi.org/10.1667/rr1409.1>
- Nerisson, P., Simonin, O., Ricciardi, L., Douce, A., & Fazileabasse, J. (2011). Improved CFD transport and boundary conditions models for low-inertia particles. *Computers and Fluids*, 40(1), 79–91.

- <http://doi.org/10.1016/j.compfluid.2010.08.013>
- O'Neill, M. E. (1968). A sphere in contact with a plane wall in a slow linear shear flow. *Chemical Engineering Science*, 23(11), 1293–1298. [http://doi.org/10.1016/0009-2509\(68\)89039-6](http://doi.org/10.1016/0009-2509(68)89039-6)
- Ono, T., Watanabe, K., Tashiro, S., Amano, Y., & Abe, H. (2017). *Experimental Studies-Fire Glove Box. Réunion NRA/JAEA/IRSN. 16/11/2017.*
- Ouf, F.-X., Delcour, S., Azema, N., Coppalle, A., Ferry, L., Gensdarmes, F., ... Yon, J. (2013). Contribution to the study of particle resuspension kinetics during thermal degradation of polymers. *Journal of Hazardous Materials*, 250–251, 298–307. <http://doi.org/10.1016/j.jhazmat.2013.01.060>
- Ouf, F. X., Gensdarmes, F., Salm, F., Lacoue, J., Bauduin, E., Brunisso, A., & Coutin, M. (2015). *Mise en suspension de particules lors d'un incendie de boîte à gants : Méthodologie expérimentale de contamination de la boîte à gants analytique. Rapport IRSN\PSN-RES\SCA\2015-04.*
- Pickering, S. (1989). The resuspension of radioactive dusts in fires. *Journal of Aerosol Science*, 20(8), 1397–1400. [http://doi.org/10.1016/0021-8502\(89\)90846-X](http://doi.org/10.1016/0021-8502(89)90846-X)
- Plagge, M., Krause, U., Riva, D., Schäfer, C., & Forkel-wirth, D. (2017). An Alternative Method for Thermal Plume – Induced Aerosol Release and Deposition Calculations in Large Geometries Using fireFoam. *Nuclear Technology*, 198(1), 43–52. <http://doi.org/10.1080/00295450.2017.1291227>
- Quintiere, J. G. (1998). A Review of Experiments on the Airborne Release of Simulated Radioactive Compounds from Fire. *Fire Technology*, 34(4), 307–324. <http://doi.org/10.1023/A:1015314510914>
- Reeks, M. W., & Hall, D. (2001). Kinetic models for particle resuspension in turbulent flows: theory and measurement. *Journal of Aerosol Science*, 32(1), 1–31. [http://doi.org/10.1016/S0021-8502\(00\)00063-X](http://doi.org/10.1016/S0021-8502(00)00063-X)
- Saffman, P. G. (1965). The lift on a small sphere in a slow shear flow. *Fluid Mechanics*, 22, 385–400.
- Sarkar, T., Anand, S., Singh, K. D., Tripathi, R. M., Sarojini, P. K., Goto, D., & Nakajima, T. (2017). Simulating long range transport of radioactive aerosols using a global aerosol transport model. *Aerosol and Air Quality Research*, 17(10), 2631–2642. <http://doi.org/10.4209/aaqr.2017.01.0049>
- Seehars, H. D. (1983). Release of Pu-containing materials during a kerosene fire. *Journal of Aerosol Science*, 14(3), 446–451. [http://doi.org/doi:10.1016/0021-8502\(83\)90158-1](http://doi.org/doi:10.1016/0021-8502(83)90158-1)
- Shang, J., Flury, M., & Deng, Y. (2009). Force measurements between particles and the air-water interface: Implications for particle mobilization in unsaturated porous media. *Water Resources Research*, 45(6), 1–14. <http://doi.org/10.1029/2008WR007384>
- Van Geel, J., Magill, J., & Schmidt, H. E. (1992). A survey of aerosol research in European community programmes. *Journal of Aerosol Science*, 23, 9–13. [http://doi.org/10.1016/0021-8502\(92\)90336-T](http://doi.org/10.1016/0021-8502(92)90336-T)
- Voelz, G. L., & Buican, I. G. (2000). Plutonium and Health. *Los Alamos Science*, 26(26), 74–89.
- Wayne, D. M., Peppers, L. G., Schwartz, D. S., & DeBurgomaster, P. C. (2018). Surface area and particle size distribution of plutonium oxides derived from the direct oxidation of Pu metal. *Journal of Nuclear Materials*, 511, 242–263. <http://doi.org/10.1016/j.jnucmat.2018.08.056>
- Wayne, D., Martinez, C., Bluhm, B., Gallimore, D., Dyke, J., D., S., & Neuman, A. (2008). *Characterization of plutonium oxide powders.*
- Yoschenko, V. I., Kashparov, V. A., Protsak, V. P., Lundin, S. M., Levchuk, S. E., Kadygrib, A. M., ... Tschiersch, J. (2006). Resuspension and redistribution of radionuclides during grassland and forest fires in the Chernobyl exclusion zone: Part I. Fire experiments. *Journal of Environmental Radioactivity*, 86(2), 143–163. <http://doi.org/10.1016/j.jenvrad.2005.08.003>
- Zepper, E. T., Brown, A. L., Pierce, F., Voskuilen, T., & Louie, D. (2017). Contaminated Fuel Fires : Parametric Sensitivity of Resuspension and Boiling Particle Evolution, 1–15.