From plasma facing components to airborne radioactive dust: A study on the behavior of tritiated dust in fusion reactors

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Nuclear fusion reactors based on plasma magnetic confinement and tokamak technology produce toxic and/or radioactive metal (beryllium and tungsten) dust as a result of plasma/wall interactions within the vacuum vessel (VV). In addition to being a factor of pollution of the plasma, these dust also involve safety issues in case of opening of the machine, maintenance or accidental situation like a break of the confinement barrier. Thus, it appears necessary to assess the adhesion and resuspension properties of these dusts during the lifetime of the reactors. To answer these safety issues, we adopted complementary approaches based on experimental and numerical works that we present in this paper.

1. INTRODUCTION

In the framework of the construction of the future ITER fusion reactor, it is important for IRSN to develop the knowledge allowing to carry out the safety expertise of this nuclear facility. To assess the safety of nuclear installations and the relevance of the associated radiation protection measures, an important step consists in determining the source terms of contamination in normal operation and for different accidental scenarios. In the event of contamination in the form of dust or aerosol, these source terms are calculated using resuspension coefficients which relate the quantity of particles emitted to the initial quantity involved, depending on the scenario considered. Under normal operating conditions of the ITER reactor, a few hundred kilograms of dust containing beryllium (Be) and tungsten (W) are expected to be produced as a result of the erosion of the walls of the vacuum chamber by the plasma. This dust will be subject to the conditions present in the reactor and may therefore be radioactive (tritium and activation products) or exhibit acute chemical toxicity (beryllium), or form, with air or water vapour, a potentially explosive mixture.

The mobility of this harmful dust and its containment in the event of an accident have become, over the years, a major subject for the safety assessment of the ITER facility. However, the behaviour of these particles remains difficult to assess, in particular due to a lack of knowledge on the evolution of adhesion forces between the particles and the surfaces on which they are deposited. In addition, radioactive dust can electrically self-charge under the effect of ionizing radiation from the radionuclides it incorporates. The resulting charged particles can be subjected to electrostatic forces that may affect their adhesion or behaviour once in the aerosol phase.

The study of the adhesion forces and the resuspension of metallic particles incorporating tritium deposited on surfaces representative of a tokamak environment are the main objectives of the work presented in this paper. In section 2, we recall the dust production mechanisms encountered in a tokamak environment. In section 3, we will introduce the experimental device and method used to sample dust inside an operating tokamak. Such work has been recently carried out in the WEST tokamak operated by the CEA in Cadarache and the dust collected allowed us to manufacture similar powders for laboratory experiments. These experiments are presented in section 4 and concern: (i) the measurement of the adhesion forces between micrometric tungsten particles and tungsten surfaces of different roughness; (ii) the measurement of resuspension coefficients when these particles are loaded with tritium and exposed to airflows having friction velocities similar to those expected in the case of a LOVA (Loss of Vacuum Accident). Results of these experiments allowed us to adapt a resuspension model in a numerical code in order to estimate the consequences of a LOVA event in a toroidal geometry. The results of these simulations will be presented in section 5.

2. ORIGINS OF DUST IN TOKAMAKS

The mechanisms responsible for the erosion of plasma facing materials can be physical or chemical and lead, under certain conditions, to the production of fine metal particles. These mechanisms are summarized in Figure 1 and correspond to: continuous sputtering by high energy particles (ions, impurities, alpha particles and neutrons); fusion and evaporation by thermal shock due to the appearance of electric arcs; blistering of surfaces by a flow of charged particles (He^{2+}, H^{+}); thermal erosion caused by strong heat flux that can detach co-deposited layers of material. Some examples of particles created by these mechanisms and found in various tokamaks are also given in Figure 1.

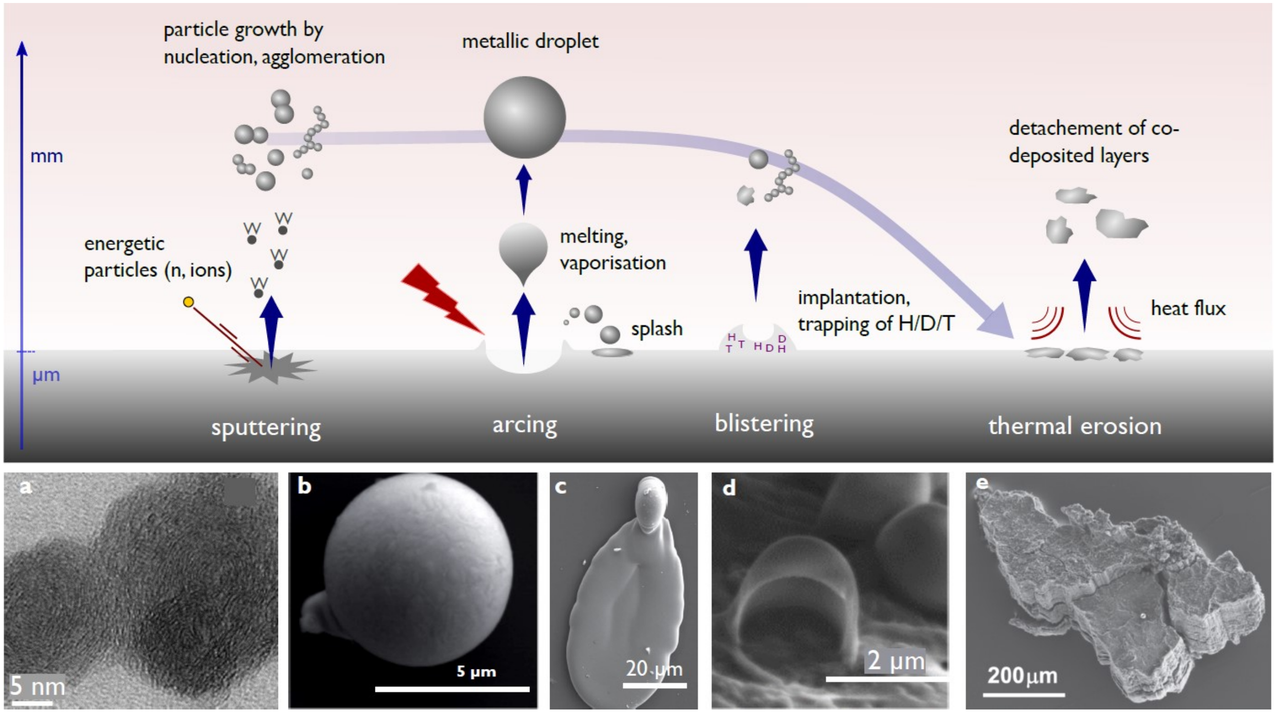

Figure 1 – (Top) Main mechanisms of dust production by plasma-wall interactions after Wirth et al. (2015). (Bottom) ***a.*** *Graphite nanoparticles collected in MAST (Arnas et al., 2010);* ***b.*** *Spherical beryllium particle from JET and* ***c.*** *Splash of beryllium in JET (Fortuna-Zaleśna et al., 2017);* ***d.*** *Bubble formation on beryllium targets in JET (Rubel et al., 2018) and* ***e.*** *Large flake retrieved from TEXTOR (Ivanova et al., 2009).*

These different dust production mechanisms are distinguished in particular by the size and specific surface area of the particles created. In fact, the chemical reactivity as well as the retention of tritium are closely linked to the specific surface area of the particles (Bernard *et al.*, 2019). The tritium inventory in the dust produced in a tokamak such as ITER will therefore strongly depend on the dominant production mechanism. Since two decades, a growing number of studies dealing with dust in tokamaks can be found in the literature and most of them have evidenced the presence of micrometre spherical particles produced by arcing. Such mechanism seems to be always present in metallic-wall tokamaks as confirmed by our recent sampling campaign in the WEST tokamak which is presented in the following section.

3. DUST SAMPLING IN A TOKAMAK

In 2018 our team has used a small sampling device named Duster Box (see Figure 2) to collect dust by controlled airflows in the WEST tokamak. The advantage of this collection technique is to investigate various locations of the plasma facing components and to provide data corresponding to particles that can be detached by airflow. A detailed description and principal characteristics of the device as well as the sampling procedure can be found in (Peillon *et al.*, 2020).

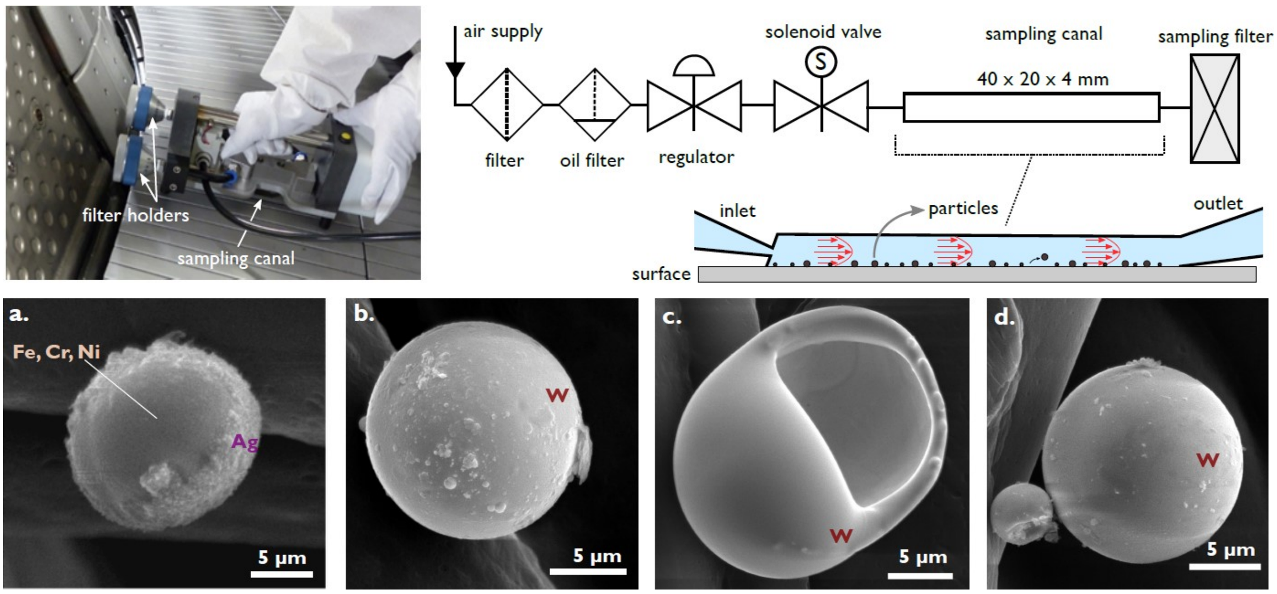

Figure 2 – (Top) Picture of the Duster Box device used in WEST and its aeraulic diagram reproduced from (Peillon et al., 2020). (Bottom) SEM micrographs showing examples of particles collected during the sampling campaign.

Whatever the probed location, spherical particles of tungsten, stainless steel, molybdenum and silver as well as dust of irregular shape coming from the coating delamination were observed. In total, 58 particles were observed with SEM and 24 of them show a spherical geometry. The typical size of spherical particles resulting from the emission of molten material droplets is in the range 5 – 30 µm. We focused our study on these particles because this size range corresponds to the inhalable fraction. SEM micrographs of **Erreur ! Source du renvoi introuvable.-a** present a spherical stainless steel particles with a coating made of silver nanoparticles. **Erreur ! Source du renvoi introuvable.-b**, 2-**c** and 2-**d** show typical tungsten spherical dust with diameters between 10 – 15 µm. Figure 2-**c** presents a tungsten bubble of approximately 22 µm likely coming from the tungsten boiling. Such bubbles have also been observed in JET but with smaller sizes (Rubel *et al*., 2018). Although the amount of particles collected during the sampling campaign was very low, these results enabled us to manufacture a surrogate tungsten powder for laboratory experiments that are presented in the next section.

4. LABORATORY EXPERIMENTS

4.1 Direct adhesion measurements

We first focused our study on the measurement of adhesion force (or pull-off force) between hard spherical tungsten particles with sizes in the micrometre range and tungsten surfaces with various roughness using Atomic Force Microscopy (AFM). Adhesion force measurements were carried out with a Multimode 8 (Bruker™) AFM in PeakForce Quantitative Nano-Mechanical mode (PF-QNM) in environmental conditions. Details of the experimental procedure and results analysis can be found in Peillon *et al*., (2019). An important experimental effort has been made to obtain spherical tungsten particles ranging from 1 µm to 20 µm in diameter thanks to a spheroidization technique introduced by Jiang and Boulos, (2006). These particles were glued on CP-FM (Colloidal Probe Force Modulation) tip-less cantilevers. Similarly, ideal smooth tungsten substrates were structured by mechanical polishing. In addition, a tokamak-relevant tungsten substrate was obtained by helium plasmas using radio-frequence hollow cathode discharges (Stancu *et al*., 2017). The root mean square (RMS) roughness of this latter substrate is 700 nm. For comparison, the surface roughness of the WEST tokamak divertor ranges between 800 nm and 1 µm (Gauthier *et al*., 2013).

The average roughness of these surfaces was determined by AFM and classified in three categories: smooth (RMS ≈ 10 nm), intermediate (RMS ≈ 200 nm) and plasma-exposed (RMS ≈ 700 nm) roughness. Examples of profiles obtained by AFM on these three samples are depicted in Figure 3 together with adhesion force measurements for a 10 µm diameter tungsten particle.

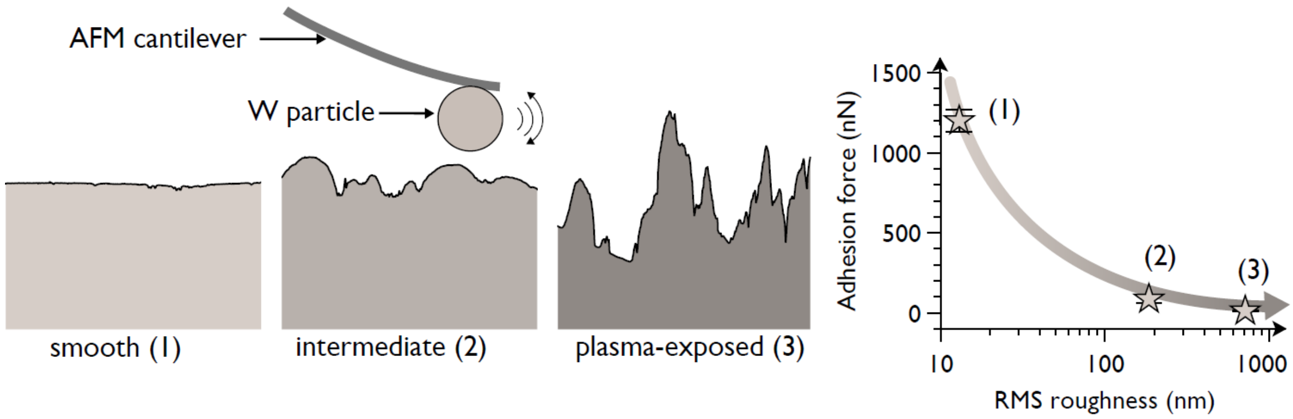

Figure 3 – (left) Profiles of the three tungsten surface roughness measured by AFM. (right) Evolution of adhesion forces for a 10 µm diameter tungsten particle with respect to the RMS surface roughness.

For a 10 µm diameter tungsten particle, experimental adhesion measurements performed with the three tungsten substrates exhibit mean adhesion forces between 1200 nN for the smooth surface to 15 nN for the rough plasma-exposed substrate. For such tokamak relevant surface, a decrease in adhesion strength of two orders of magnitude in comparison with the ideal case (smooth surface) has been evidenced. Moreover, we have found that for surface roughness greater than RMS ≈ 100 nm, the size of the particles becomes a secondary parameter in the evolution of adhesion forces. Until now, such feature was not quantitatively considered in resuspension models where roughness of the substrate is generally taken as an adjustment parameter. A similar AFM study has been performed with a glass substrate (not shown here) and we demonstrate in the following paragraph that such a study can be successfully used, together with a resuspension model, to describe the resuspension behaviour of a tungsten powder deposited on a flat glass substrate.

4.2 Resuspension experiments

Following adhesion measurements made with AFM, we have implemented resuspension experiments with tungsten powders in order to validate the resuspension model of Reeks and Hall (2001) for its use in a numerical code. These experiments were performed with a tungsten powder composed of spherical particles of 10 µm in diameter deposited on a glass substrate with RMS roughness of 5 nm. In order to assess the influence of tritium retention in the resuspension of such dust, a batch of the powder has been tritiated in a dedicated nuclear glove box at CEA Saclay. The specific activity obtained for such tungsten powder is 300 $MBq.g^{-1}$. The resuspension experiments were conducted in a glove box using the Duster Box as a miniature wind tunnel. Resuspension coefficients were obtained by means of optical microscopy using a *Morphologi G3* Malvern microscope. This apparatus allowed us to count the number of particles present on the substrate before and after their exposure to airflows with various friction velocities.
The resuspension coefficients (KR), corresponding to the ratio of detached particles over the initial amount of particles present on the substrate, are reported in Figure 4 in terms of airflow friction velocity and for the two powders (stock and radioactive powder). Resuspension model calculations using AFM force distributions previously obtained are represented by the continuous blue line in Figure 4. Taking into account the standard deviations found for these measurements, one can consider that the resuspension coefficients between the tritiated and non-tritiated powders are equivalent. This result is not surprising given the low specific activity found for these micrometre tungsten particles. Indeed, for such particle size and specific activity, we have calculated that the electrostatic image force is in the order of 10^{-3} nN, thus playing no role in the adhesion of the particles (Peillon *et al.*, 2020).

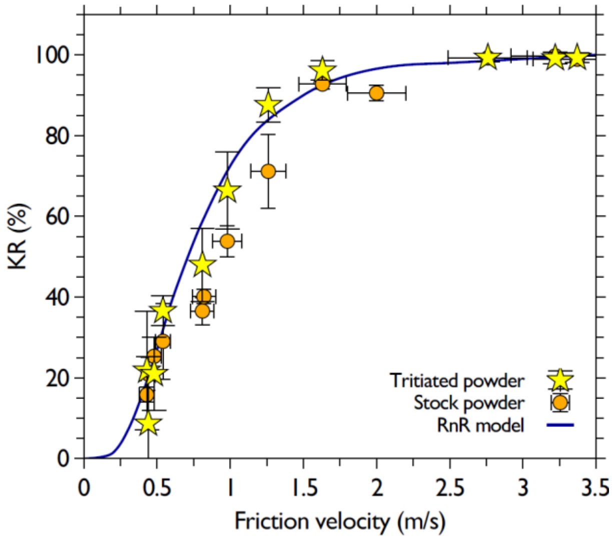

Figure 4 - Resuspension coefficient (KR) obtained for stock tungsten powder and tritiated tungsten powder according to the airflow friction velocity. The Rock'n roll model calculations, integrating adhesion force distribution obtained by AFM, is depicted by the continuous blue line.

On the other hand, thanks to the adhesion force distribution obtained with the AFM measurements, we were able to obtain a very good agreement between the analytical Rock'n roll resuspension model of Reeks and Hall (2001) and resuspension experiments performed with tungsten powders. Therefore, we have implemented this model in a numerical code in order to evaluate the resuspension of particles in a tokamak-like geometry. This work is introduced in the next section.

5. THEORETICAL MODELLING AND SIMULATION

To evaluate the resuspension of particles from the airflows generated during a LOVA, numerical simulations were performed using *ANSYS CFX* software, into which the resuspension model of Reeks and Hall (2001) was implemented. The first step consists in representing the geometry of the torus with the duct that simulates the breach, which is the source of the loss of vacuum. For the computational domain, a toroidal geometry of 1300 m^3 is retained with a D-shaped section. To represent the break, a circular duct with a diameter of 0.16 m and a length of 2 m is used. The duct is located halfway up the torus and is initially closed at the torus wall. Single spherical tungsten particles of 10 µm in diameter, represented in yellow in Figure 5, are deposited at the bottom of the torus (made of tungsten) before simulation starts.

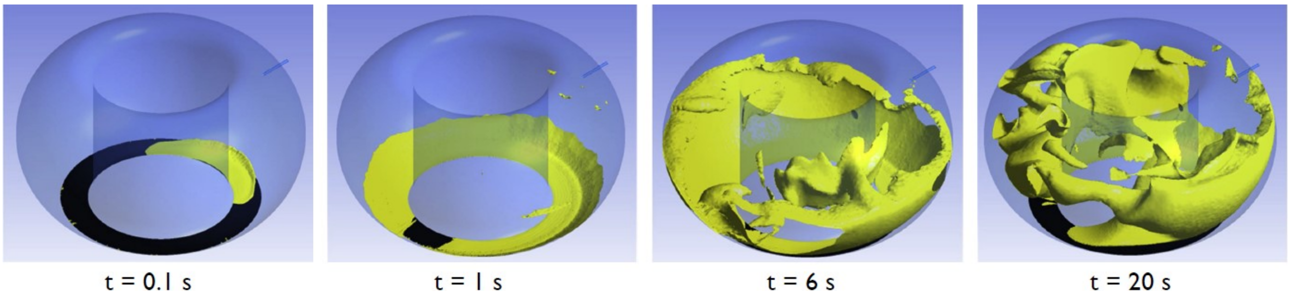

Figure 5 - Time evolution of the resuspension of 10 µm tungsten particles in case of a LOVA in a toroidal geometry (Gelain et al., 2020).

The resuspension phenomenon occurring during the calculation is illustrated in Figure 5 which tracks the evolution of the resuspension of particles and their dispersion during the pressurization. Mean friction velocities at the bottom of the torus are between 1.5 $m.s^{-1}$ and 4 $m.s^{-1}$ just after the breach. At the beginning (t = 0.1 s), the friction velocity is maximum (≈ 8 $m.s^{-1}$) close to the air entry and below the impaction with the wall which initiates resuspension of particles located in this area. At the time 1 s, the friction velocity is high on a larger area of the floor. For the following times 6 s and 20 s, the friction velocities on the floor are lower, so the resuspension too, and what is observed is mainly the particle dispersion in the torus. Such calculations allow to track the kinetics of resuspension and to obtain the resuspension coefficient as a function of time. At the end of the simulation, approximately 10 % of the 10 µm diameter particles have been resuspended. Hence, for an initial deposit of 1000 kg of tungsten dust (safety limit for ITER is 1 kg of tritium and 1000 kg of dust according to Taylor and Cortes, 2014), about 10 kg of tungsten dust would be in aerosol phase inside the reactor. If the aerosol is evenly distributed in the chamber, this corresponds to a dust concentration of 7 $g.m^{-3}$, well below the explosibility limit for tungsten dust (above 500 $g.m^{-3}$ according to Denkevits and Dorofeev, 2006). These simulations demonstrate the feasibility of particle resuspension CFD simulations with a resuspension model taking into account true description of adhesion forces. Moreover, from a safety point of view, these simulations allow one to obtain the final concentration of particles in aerosol form inside the reactor which can be used to assess the dust concentration explosion limit or tritium inventory limit inside the vacuum vessel.

6. SUMMARY

In the present work, collection of particles inside the WEST tokamak has been realized thanks to a new *in-situ* particle collection system called Duster Box. This method is innovative and allowed to identify various types of tungsten particles among them spherical shaped micro-particles in the micrometre range. Moreover, a detailed description of adhesion forces between tokamak-relevant particles and substrates has been performed. For this purpose, a parametric study using Atomic Force Microscopy (AFM) was completed and adhesion forces were obtained for spherical tungsten particle of 10 µm in diameter and three tungsten substrates with RMS roughness between 10 nm and 700 nm, the latter representing usual roughness of tokamak surfaces in contact with plasma. This technique made it possible to validate an analytical resuspension model that was successfully integrated in a resuspension numerical code. Thanks to its simplicity and small dimensions, the Duster Box device has also been used for resuspension experiments in a nuclear glove box with tritiated particles. The feasibility of such experiments has been demonstrated and no differences in the resuspension were observed between neutral or tritiated particles. The results of these experiments combined with the implementation of the particle removal model in a CFD simulation provide robust data for the determination of the tritium source term which is mandatory for the definition of workers radioprotection plans. Moreover, such data are essential when assessing the dispersion of toxic/radioactive material in the environment that could follow a loss of containment.

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