Phenomenological modelling of particle resuspension on bubbling PMMA.

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Along the nuclear fuel industrial process, radioactive materials are handled in confined glove boxes, which retain a surface contamination by radioactive solid particles. Evaluating the potential release of radioactive particles in case of a fire occurring in a nuclear facility is of critical interest for nuclear safety studies. Several experiments have been conducted in order to gather reference data for these studies, highlighting a specific behaviour of PMMA, namely a flash of emission. This flash is often related to the bubbling of PMMA, but so far no experimental proof has been obtained to support this allegation.

A previous study proposed an empirical correlation between the incoming heat flux and the onset of the flash (Ouf *et al.*, 2013). One can note that this correlation fits with the time to reach a surface temperature of 275°C, which is the starting temperature for thermal degradation of PMMA, according to thermo-gravimetric analysis. At 275°C, the depolymerisation process starts, yielding monomer molecules (Methyl methacrylate – MMA). MMA's boiling temperature is around 100°C, so the pyrolysis products are overheated and will immediately nucleate into bubbles. This observation supports the previous assumption but a more detailed investigation is required.

An experimental study of the bubbling of PMMA is conducted to gather specific data on the bubbling, in order to validate a theoretical model. Three pristine PMMA samples are exposed to a heat flux of 45 kW/m^2, using a radiant panel (modified cone calorimeter), for durations of 40, 50 and 60 seconds and then quenched in water to stop the thermal degradation. X-ray tomography is then performed on the bubble layer and a three dimensional reconstruction is performed before analysing the bubble layer's microstructure.

Bubble size distribution (volume equivalent diameter) and size-to-depth profiles are obtained for all three samples (see figure 1 for the 60 s sample).

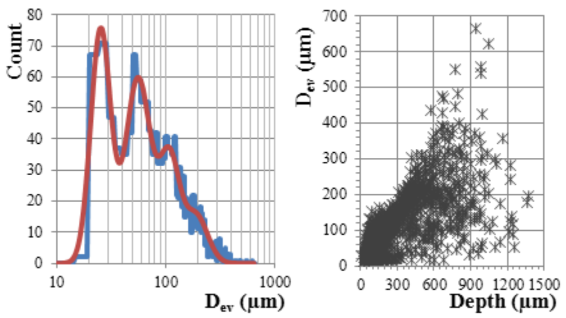

Figure 1. Bubbles size distribution and size-to-depth profiles inside PMMA exposed during 60 s to 45 kW/m^2

An increase in mean size with depth is observed with all three samples. This indicates that the bubbling-induced resuspension is mostly caused by the large amount of small (< 200 µm) bubbles that burst in the early phase of thermal degradation; probably because of their higher internal pressure.

A physical model of resuspension of a solid particle by a bursting bubble is proposed (see figure 2), based on the pressure difference between the bubble and the atmosphere.

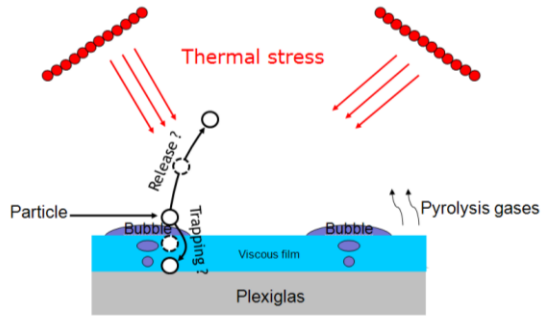

Figure 2. Modelling of the bubble-induced resuspension

This pressure difference is obtained using the ThermaKin code (Stoliarov and Lyon, 2008) to obtain a full thermal model of the polymer sample, including the effects of degradation; the pressure difference is then computed using the Laplace law with a model of surface tension. Effects of drag and inertia are taken into account in a one-dimensional, mechanical model. This model is then refined to include the surface density of the particle deposit and the bubble bursting frequency to obtain a value for the flux of particles. No hypothesis is made on the nature of the particle, allowing us to investigate radioactive particles (PuO_2).

Further developments of this model will include the effects of gravitational and thermal migration (Soret effect) of the particles inside the viscous film of polymer. These phenomena will tend to trap the particles and block their release, acting oppositely to the bubbling.

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