

# Study of the formation of aerosols of FPs (I, Cs, Mo) and control rod material (Cd) in conditions representative of the RCS during a severe accident and consequences on gaseous iodine release

A.C. Gregoire<sup>1</sup>, E. Assaf, S. Sobanska<sup>2</sup>,

<sup>1</sup>Institut de Radioprotection et de Sûreté Nucléaire, IRSN, BP 3, F-13115 St. Paul lez Durance Cedex, France

<sup>2</sup>Laboratoire de Spectrochimie Infrarouge et Raman, UMR CNRS 8516, Université de Lille, Sciences et Technologies, 59655 Villeneuve d'Ascq Cedex, France

Keywords: Pressurized water reactor, Severe accident, Aerosol formation, Iodine release

Presenting author email: anne-cecile.gregoire@irsn.fr

In a nuclear reactor severe accident (SA), the volatile fission products (FPs), the structural and control rod materials released from the degraded core are transported and partly deposited in the reactor coolant system (RCS). During their transport to the containment, they undergo physical and chemical processes leading to the formation of aerosols and gases (Haste and Payot, 2013). The nature and concentration of these species which are then injected in the containment will strongly determines the potential source term to the environment, especially for iodine, which is a crucial issue for accident management.

Indeed, even if CsI is from a thermodynamical point of view very stable, some elements (Mo, B) can act as a caesium sink, preventing CsI formation and contributing to the release of gaseous iodine early during the degradation phase (Girault and Payot, 2013).

Experimental studies on the behavior of FPs in the reactor coolant system have been investigated for many years at IRSN. The CHIP experimental program (Chemistry Iodine Primary Circuit, Clément and Zeyen, 2005) is dedicated to obtain information both on iodine speciation and aerosol composition under different circuit boundary conditions in presence of other FPs (Cs, Mo) and/or control rod materials (Ag, In, Cd, B).

The influence of Mo was firstly studied in a laboratory-scale test line with concomitant injection of CsI and MoO<sub>3</sub> vapours in steam conditions (Gouello *et al.*, 2013). The formation of caesium molybdates condensed species was observed even with low amounts of Mo. Complementary experiments were performed in the CHIP PL facility (Grégoire *et al.*, 2015) with injection of I<sub>2</sub>, CsOH and MoO<sub>3</sub> vapours, allowing elemental ratio representative of fission products transport in the RCS in SA conditions (excess of Mo relative to Cs and of Cs relative to I). The influence of the atmosphere was explored with tests performed in oxidizing conditions (steam) and with tests performed with more reducing conditions (mixture of steam and hydrogen). Finally, the influence of Cadmium was also evaluated through the additional injection of Cd(0).

In this work, the deposits collected in the 700-400 °C temperature range and aerosols transported to the line outlet (150 °C) were characterized for their size, morphology and elemental composition by Scanning Electron Microscopy coupled with an Energy Dispersive

X-ray spectrometer (SEM-EDX). The molecular composition of the heterogeneous mixtures was determined by combining, X-rays diffraction and Raman microspectrometry. Finally, the analysis with X-rays photoelectron spectrometry (XPS) allowed determining the surface composition and the oxidation state of elements.

Experimentally it has been shown that in oxidising conditions, caesium can be completely consumed to form dicaesium polymolybdates, promoting thus the formation of gaseous iodine. The particles collected at 150 °C are submicrometer needle like particles. I<sub>2</sub> adsorbed at the particle surface was evidenced too. In reducing conditions, CsI remains the dominant form of iodine, as the amount of oxygen is not sufficient to allow formation of quantitative caesium polymolybdates. Reduction of Mo(VI) into Mo(IV) also explained the limited formation of polymolybdates species. Again submicronic particles were collected at 150 °C. Finally, the presence of Cd induced a competition between Cd and Cs in molybdate-rich particle formation, reducing the amount of released gaseous iodine.

Both, formation of submicronic particles and release of gaseous iodine may be of concern in case of severe nuclear accident.

This work was supported by funds from the “Laboratoire d’Excellence” (LABEX) -CaPPA- (ANR-11-LABX-0005-01) and CHIP experimental program partners.

Clément, B., Zeyen, R., (2005) In: *Proceed. of International Conference on Nuclear Energy for New Europe 2005*. Nuclear Society of Slovenia, Bled, Slovenia, 5-8 September.

Girault, N., Payot, F., (2013). *Ann. Nucl. Energy*, **61**, 143–156.

Haste T, Payot, F., Bottomley, P.D., (2013), *Ann. Nucl. Energy*, **61**, 102–121.

Gouello, M., Mutelle, H., Cousin, F., Sobanska, S., Blanquet, E. (2013) *Nucl. Eng. Des.* **263**, 462–472.

Grégoire, A. C., Kalilainen J., Cousin F., Mutelle H., Cantrel L., Auvinen A., Haste T., Sobanska S. (2015), *Ann. Nucl. Energy*, **78**, 117-129.