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Title page

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Study of the radiolytic decomposition of CsI and CdI₂ aerosols deposited on stainless steel, quartz and Epoxy painted surfaces

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Study of the radiolytic decomposition of CsI and CdI₂ aerosols deposited on stainless steel, quartz and Epoxy painted surfaces

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Abstract

CsI and CdI₂ aerosol decomposition rate under irradiation has been quantified at 80°C and 120°C in presence of humidity and on different substrate (stainless steel, quartz and Epoxy paint). A model has been developed for the ASTEC-SOPHAEROS code to reproduce the data and help the identification of the gaps remaining in the understanding of iodine volatility in a severe accident of a Nuclear Power Plant (NPP). The current model applied to model the gaseous iodine behaviour in the containment of PHEBUS-FP tests does not fit with the experimental data probably because the nuclear aerosol reaching the containment are much more complex than pure CsI aerosols. It has been clearly shown than the radiolytic oxidation of metallic iodide aerosol into molecular iodine can significantly impact the source term evaluation even if additional experimental data area required to cover the variety and complexity of nuclear iodide aerosols.

Keywords

Iodine, Caesium iodide aerosol, Cadmium iodide aerosol, Radiolytic decomposition, Kinetics, Severe accident, PHEBUS-FPT tests

Introduction

In case of severe accident (SA), iodine is one of the most hazardous fission products (FP) that would be released from the fuel to the reactor coolant system (RCS) and then to the containment of a nuclear power plant. Once in the containment, iodine species undergo physical and chemical phenomena (including thermal and radiolytic reactions in different media like the sump, the gaseous phase and the surface interactions). The balance between these phenomena determines the amount of iodine that could be released in the environment due to the containment leakages or venting. For about 40-50 years, chemical reactions having a significant influence on iodine volatility have been

identified and their kinetics has been quantified, modeled and capitalized in SA codes like ASTEC [1,2,3,4,5]. The sump and gaseous species in the containment have been well identified (I₂, organic iodides like CH₃I, AgI, HOI, I, IO₃, and more recently iodine oxides aerosols) [6,7,8,9,10,11,12,13]. However, an uncertainty remains on the speciation of iodine entering into the containment from the RCS. In fact, once they are released from the nuclear fuel at temperatures ranging from ~ 1500 to 2500 K (for which some FPs could be on their atomic form), FPs are expected to recombine in the RCS as the temperature decreases [14,15,16] down to 373-473 K. The speciation of iodine entering into the containment has largely been debated in the nuclear safety community. Nevertheless, PHEBUS-FP tests [17,18,19,20,21] have shown that iodine can be under two physical forms once it arrives into the containment: gaseous and aerosols. Even if the gaseous species is expected to be mostly inorganic (I2 and/or HOI/HI) [22,20] the speciation of iodine aerosols is still being debated. In the RCS, Iodine and Caesium have been assumed for several decades to be bounded (CsI species) [23]. In fact, from thermodynamic calculations [24,25,26], experiments [27] and the ratio between Cs and I masses transported in different kinds of RCS experiments [28,29,30], it has been assumed that CsI could be the main aerosols species entering into the containment [31,32,33]. CdI₂ aerosol has also been identified [34,35,36] and its importance has been assessed in more recent studies [37,38,39]. As soluble species, CsI and CdI₂ aerosols lead to the formation of iodides ions as soon as they reach the aqueous sump and could play a significant role on iodine volatility (under irradiation, it is well known that I ions can be oxidized into volatile molecular iodine). However, many other FPs and structure materials (Ag, In, Cd, B) are also transported in the RCS in significant amount and can react with iodine. They could participate to the iodine aerosols composition as they are not necessarily soluble in the sump. This is supported by the PHEBUS-FPT-1 test [18] for which the aerosols composition in the RCS has been found to be more complex than expected. In fact, it has been found that the aerosols are multicomponent, containing structure material and FPs (like oxidized silver, Ag, In, Cs, Sn, U, Cl, C, O, Ni). Iodine could be bonded not only to Cs but also to Ag (or oxidized silver) for example and could lead to insoluble aerosols (or partly insoluble) reaching the containment as observed in PHEBUS FPT-0, FPT-1 and FPT-2 [17,18,19]. In the containment, these iodine multicomponent aerosols also settle down on dry surfaces. However, their stability under irradiation is not known. They might be stable but the irradiation field could participate to their decomposition and lead to the formation of gaseous iodine. In fact, from the XPS analysis of PHEBUS FPT1 aerosols [18], iodine atoms might be expected to be located in the outer and inner shells of these multicomponent aerosols and to be bounded to other FP and structure material which makes it difficult to anticipate and predict if such deposited complex aerosols would be stable under irradiation. Moreover, even if significant progresses have been made for 20 years in understanding and modeling iodine behavior in the containment, there are still significant uncertainties in the estimation of iodine volatility [40]. It is

thus needed to better quantify the unknown processes that could participate to the iodine volatility. especially in the gaseous phase and at the interface with surfaces. As iodine volatility in PHEBUS-FP tests is still significantly underestimated by the current models when deposited iodine aerosols are considered stable under irradiation [40] (i.e assuming they are not decomposed into inorganic iodine), some efforts have been put lately on the study of the stability of iodine oxides aerosols (IOx) under irradiation and of multicomponent iodine aerosols deposited on different kind of surfaces (stainless steel, quartz and Epoxy paint) within the OECD/STEM and STEM2 projects. The main objective is to check whether iodine aerosol decomposition could participate significantly to iodine volatility or not. The study of IOx decomposition under irradiation is addressed in a separate article. CsI and CdI₂ aerosols are widely cited in the literature as the main species being released in the RCS and then into the containment [28,29,30,31,32,33,34,35,36,37,38,39]. Despite the fact that CsI behavior (and CdI₂ in a lesser extent) has often been studied in the sump or at high temperature in the RCS [41,42,43,44], or under UV radiation to look at the effect on its agglomeration properties [45,46], there is no record in the literature about its decomposition into volatile iodine under irradiation at temperatures representative of the containment (< 200°C). We have thus studied the decomposition of both aerosols first. Both are soluble aerosols which is quite practical for manufacturing and tracing these aerosols with ¹³¹I which is necessary to investigate their decomposition under irradiation in the EPICUR facility. This paper gives an overview of the experiments performed in this area and the related results. A second part of the paper deals with the interpretation of these data with the ASTEC-SOPHAEROS code (V2.1) [5]. A model was optimized from the experimental data and applied to the modeling of PHEBUS-FPT tests in order to assess the importance of such aerosols decomposition towards iodine volatility. A discussion is finally made to conclude on the phenomena that still remain to be studied in order to better predict iodine volatility in case of a severe accident.

1 Description of STEM experiments in the EPICUR facility

1.1 Experimental set-up

The experimental set-up used to study the radiolytic decomposition of CsI and CdI₂ aerosols under radiation consists of a loop containing a panoramic irradiator, an electro-polished stainless steel irradiation vessel (4.8 l), connected through electro-polished stainless steel tubes to an iodine filtration system, called Maypack (**Fig. 1**). The panoramic gamma-ray irradiator (6 sources of ⁶⁰Co delivering an average irradiation dose rate of several kGy.h⁻¹) is used to simulate the effect of radiation associated with the presence of radioactive fission products in the containment vessel during an accident.

After the iodine aerosols loading phase on the coupon (see section 1.2.2), the coupon is installed in the temperature-controlled irradiation vessel connected through stainless steel tubing to the Maypack in the EPICUR facility. The different volatile iodine species released from the CsI or CdI₂ aerosols during the tests in the irradiation vessel are transferred to the May-pack device and trapped on the different species-selective filters: the quartz filter (QF) traps the aerosols species, Knit-mesh filters (KM) trap molecular iodine and charcoal filters (CA) trap others species, particularly organic iodides in our experiments. Thanks to the use of ¹³¹I labelled CsI and CdI₂ aerosols (see section 1.2.2), gamma measurements are performed placing a gamma counter above each stage of filter of the May-pack device, allowing on-line measurements and the evaluation of the release kinetics of the different volatile species during the test.

At the end of the tests, the irradiation vessel and the May-pack device are recovered. The different filters of the May-pack, the coupon and the rinsing solutions of the irradiation vessel of the experimental loop are also gamma counted in order to compare these post-test measurements with the on-line measurements, check the activity balance and evaluate the missing activity balance (MAB) at the end of the tests.

1.2 Samples preparation

1.2.1 Preparation of the coupons

Three kinds of coupons were used for these tests: painted, stainless steel and quartz coupons. The epoxy painted coupon were made of black steel, recovered with 2 layers of CENTREPOX® (40 μ m) and 1 layer of HYDROCENTRIFUGON® (50 μ m). HYDROCENTRIFUGON® paint is an epoxy type paint. It was chosen as this type of paint is often used to cover the inner containment surfaces of Pressurized Water Reactor (PWR). CENTREPOX® is also an epoxy type paint used to avoid the corrosion of the containment surfaces. The fresh epoxy painted coupons were preheated at 130°C for 96 h in a dry atmosphere prior to the loading phase and the irradiation phase (a conditioning procedure to ensure a representative paint state surface for iodine adsorption [47]). The stainless steel and quartz coupons were cleaned with demineralized water, then with alcohol, then dried with Joseph paper to remove all traces of grease. No other treatment was performed on these coupons before the loading and irradiation phases.

The aerosols loading phase consist in using an aerosols generator (AGK2000 from PALAS®) including a spray nozzle, a cyclonic separator and a dryer in order to convert CsI or CdI₂ solutions to solid aerosols. The generated aerosols were deposited by gravity and were uniformly distributed on the total surface of a disk-shaped coupon, on one face of the coupon (24.6 cm²).

The CsI deposited concentration on the coupon were set up considering that the total iodine core inventory of a NPP would be released in the containment under CsI/CdI₂. Given the ratio between the horizontal and vertical surfaces, and based on the ratio of aerosols that have been deposited (\approx 65%) and diffused on the vertical walls (\approx 2%) of PHEBUS containment, the maximum CsI deposited concentration ranges between 10⁻³ – 10⁻² mol.m⁻² for horizontal surfaces and between 10⁻⁵ – 10⁻⁴ mol.m⁻² for vertical surfaces. These orders of magnitude were considered to set up appropriate CsI and CdI₂ concentration in the aqueous solutions. As CsI and CdI₂ are soluble in aqueous solutions, solutions were prepared using commercial CsI or CdI₂ powders (Sigma-Aldrich®) solubilised in water to get a concentration of 120 g.l⁻¹ (to reach an aerosols concentration of ~10⁻⁴ mol(I).m⁻² on the coupon) or 10 g.l⁻¹ (to reach an aerosols concentration of ~10⁻⁴ mol(I).m⁻² on the coupon) or 10 g.l⁻¹ (to reach an aerosols concentration of ~10⁻⁴ mol(I).m⁻² on the coupon). Na¹³¹I was added to the solutions in order to label the CsI/CdI₂ generated aerosols. As NaI concentration is very low compared to CsI/CdI₂ concentration, we do not expect significant NaI aerosols on the coupon (for an initial CsI concentration in the solution of 120 g.l⁻¹, the Cs/Na ratio is higher than 100).

In order to check the speciation on the coupon, one coupon was washed and the molar ratio I/Cs and I/Cd was quantified with Inductively Coupled Plasma (ICP) technique. For CsI, the molar ratio was found to be ≈ 1 whereas it was found to be ≈ 2 for CdI₂. The Iodine species recovered on the coupon is thus expected to be CsI/CdI₂.

The size of the aerosols deposited on the coupons were evaluated by means of observations performed by optical microscopy of a coupon loaded with inactive CsI aerosols, using the same conditions as for labeled CsI. These observations showed that the aerosols size (GMD: Geometric Mean Diameter) was a function of the initial CsI concentration of the solution. Using a less concentrated solution led to the formation of smaller CsI aerosols with a lower concentration (GMD $< 3 \mu m$ with a concentration of 10 g.1⁻¹ versus GMD $< 9 \mu m$ with a concentration of 120 g.1⁻¹). These aerosols are larger than the PHEBUS aerosols size whose GMD is $0.5 - 1 \mu m$ [18].

The initial iodine concentration expressed in mol.m⁻² on the coupon is calculated, using the activity measured on the coupon (after gamma counting) and assuming that the ratio 127 I/ 131 I on the coupon is the same than the one in the solution. The concentrations of iodine aerosols deposited on the coupons are summarised in Table 1.

1.3 Painted coupon analysis

After the end of the test, painted coupon were first counted with gamma spectroscopy to quantify how much iodine has been volatilized. Then, it was immersed in a sodium hydroxide solution (0.1 mol.L⁻¹) for 12 hours in order to dissolve CsI and have an estimation of how much Iodine and Caesium remain bounded to the paint. The quantification was performed with ICP technique several months after the end of the test.

1.4 Experimental conditions

The experimental conditions of the iodine aerosols decomposition tests under irradiation are recalled in **Table 1** and **Table 2**. The objective of these tests was to check if CsI and CdI₂ aerosols are decomposed in representative conditions of the containment under irradiation and to quantify the decomposition rate. Each test is divided into 3 phases:

- a pre-irradiation phase from 0 to 1 hour to check the iodine release due to thermal processes.
 However, the short duration of this phase for most of the tests (except AER12) did not allow to conclude on the effect of the temperature on CsI thermal decomposition. Complementary thermal CsI decomposition tests were thus performed at 105°C (see below) to check its thermal stability for longer exposure times.
- the irradiation phase (~ 30 hours) to check the iodine release due to the irradiation of the iodine aerosols
- and the post-irradiation phase (from 4 to 10 hours) to check whether volatile iodine is still released after the irradiation is stopped.

Various parameters have been varied in order to cover different representative situations and surfaces that might be found in a containment reactor:

- as both types of aerosols are soluble, steam could have a significant influence on the aerosols stability under irradiation. Thus, the humidity influence was checked with AER1 (R.H = 20 %) and AER2 (R.H = 50 %) tests, performed with CsI deposited on a quartz coupon.
- the influence of the dose rate and initial concentration of CsI aerosols on an Epoxy paint surface was checked with AER5, AER6 and AER7 tests.
- The temperature is also an important parameter. It was thus varied between 80°C (AER6) and 120°C (AER11), performed on a stainless steel coupon.
- The aerosols nature has also been investigated to check iodine volatility from one soluble aerosol type (CsI) to another (CdI₂).

A mixture of air and humidity was used as the carrier gas in the EPICUR irradiation vessel with a fixed gas flow rate that allows a low residence time of the released iodine species in the vessel (~ 20 minutes).

1.5 General gaseous iodine release profile and associated uncertainties

A typical release profile from deposited CsI aerosols on quartz under irradiation is given on Fig. 2.

It can be seen that CsI aerosols are not stable under irradiation as a significant part of the initial iodine inventory is recovered on the Knit-mesh filter. Moreover, their decomposition leads mostly to the formation of volatile inorganic iodine (assumed to be molecular iodine [40]). A significant activity has also been observed for some tests on the quartz fibre filter (for aerosols trapping) which could be a mixture of iodine oxides aerosols (IOx) formed during the residence time of gaseous iodine in the vessel and gaseous iodine adsorbed on the quartz filter (the CsI aerosols vaporization or the mechanical resuspension are not likely in these conditions). Nevertheless, both species are inorganic iodine decomposition products from of deposited CsI aerosols.

There is also a significant activity on the charcoal filter (CA) that represents less than 5% of the total release for AER2 (and less than 15% for all the tests). Organic iodides are usually found in significant amount under irradiation when iodine is adsorbed on Epoxy painted surfaces [40] or polymers. For AER2, there is no painted coupon and thus less organic material available for reaction with gaseous iodine in the vessel and the loop. Among all the tests performed in EPICUR facility without painted coupons since the beginning of its use, such low amount of activity on the charcoal filter have always been observed. As the carrier gas also contains a small amount of organic pollutions (that could also be from the facility itself), it is likely that organics could react (through a radiolytic process) with gaseous I₂ during their residence time in the irradiation vessel and loop [40,48]. This activity could also arise from small amounts of inorganic iodine that have not been trapped by the inorganic Knitmesh stage (KM).

The precise gamma spectrometry measurement of the iodine amount on the coupon before and after the test (which gives precisely the amount of iodine that has been released) indicates that (except for paint surfaces tests and AER1 test performed at a lower R.H), 99% of the iodine inventory has disappeared from the stainless steel and quartz coupons (**Table 3**). Despite the loop and vessel washing (at the end of the test) that allowed to recover up to $\sim 33\%$ of the iodine initial inventory, there is still a significant MAB for most of the tests (from 4 to 33% of the initial iodine inventory in **Table 3**), as in previous studies [40]. It is assumed that gaseous inorganic iodine is partly lost on the stainless steel surface of the vessel and on the loop surfaces of the facility (between the irradiation vessel and the Maypack) on which it can be irreversibly adsorbed [49,59].

In order to consider the real amount of gaseous iodine that has been released from the aerosols deposited on the coupons, the raw data (example given on Fig. 2) had to be corrected for each test, mostly based on the MAB and loop and surfaces washing. Other minor corrections were also made on the raw on-line data due to (1) the knit-mesh efficiency and (2) the activity detected on the quartz filter that was assumed to be inorganic iodine. Both were added to the on-line knit-mesh data. All of these corrections are detailed and discussed more deeply in a previous paper [40]. The relative uncertainty on the on-line corrected data was estimated to be around 35% in the same paper, i.e roughly one third of the corrected on-line value.

2 **Results and discussion**

The final on-line measurements of volatilized organic iodide (RI) and inorganic iodine (I_2) fractions are presented in Table 3. The global volatilization from the quartz and stainless steel coupons is

very important (close to 100% at 50% R.H.) which shows that CsI and CdI₂ aerosols are not stable under irradiation and significantly decompose.

The influence of several parameters was checked and is discussed in the following section.

2.1 Effect of the temperature without irradiation

On Fig. 3 to Fig. 9, the effect of the thermal pre-irradiation phase (1 h and 10 h for CsI and 10 hours for CdI₂) and the post-irradiation phase (4 h) is not significant which shows that CsI aerosols seems stable when the irradiation is not set up or after it is stopped. A blank test (no irradiation) was performed to check CsI aerosols thermal decomposition over 70h (DT4) at 105°C and in humid atmosphere (50 % RH). No significant thermal decomposition of CsI aerosols was observed which indicates that CsI aerosols are stable at 105°C and lower temperatures. This is consistent with AER12 test (80°C) showing no CsI aerosols thermal decomposition during 10 hours before the irradiation starts (Fig. 4). The thermal CsI aerosols decomposition is thus expected to have been non-significant in EPICUR tests performed at 80°C. For AER11 test performed at 120°C, even if no blank test could be performed at this temperature, we do not expect a significant CsI thermal decomposition because the 1 hour thermal phase prior to the irradiation did not show any release on the Maypack and also from the results at 80 and 105°C (no release).

2.2 Effect of the temperature under irradiation

The effect of the temperature under irradiation was studied with AER6 and AER11, with CsI aerosols deposited on a stainless steel coupon. Fig. 5 shows the corrected inorganic iodine release on the knit-mesh filter versus the irradiation time. It can be observed that, despite the same pre-irradiation phase duration (1 hour), the release for AER11 (120°C) starts approximately two hours after the AER6 (80°C) release. The inorganic iodine release seems lower at 120°C than at 80°C. However, as the uncertainty on the data can be considered to be ~ 35%, it can be concluded that the temperature influence on CsI decomposition is not really significant in the 80-120°C temperature range. After 30 hours of irradiation, CsI aerosols have been almost completely decomposed into volatile iodine at both temperatures.

2.3 Effect of the relative humidity (R.H.) on CsI and CdI₂ aerosols decomposition

The effect of the humidity (20 % and 50%) on CsI aerosols decomposition under irradiation is shown on Fig. 3. A low humidity promotes a higher CsI stability, especially in the long term (> 20h)

for which a release plateau is observed (Fig. 3) whereas a 50% R.H leads to a complete CsI decomposition into inorganic volatile iodine in \sim 30 hours of irradiation. As shown on Fig. 4, it is even faster for CdI₂ as it entirely decomposed in less than 20 hours.

We expect the humidity to hydrate the soluble CsI and CdI₂ aerosols, forming a water shell around the compounds with possible formation of ionic species, I-, at the interface particule/water. On top of that, we expect two other phenomena: (1) CsI thermal decomposition by steam and (2) CsI radiolytic decomposition by steam oxidant radicals like HO^{\circ} (and also may be O^{\circ}) (reaction (1) to (3) below). The free energies estimation at 298K from thermodynamic data [50,51,52,53] are also given, considering all reactants and products as gaseous compounds. Free energies in brackets refer to reactions with Caesium compounds (reactants and products) considered as solid compounds.

(1)	Thermal process :	CsI	+	H_2O	\leftrightarrow	CsOH +	HI	$\Delta G_r^{\circ}(298K) = +166 kJ.mol^{-1} (+189)$
(2)	Radiolytic processes:	CsI	+	HO°	\leftrightarrow	CsOH +	I°	$\Delta G_r^{\circ}(298K) = -28 \ kJ.mol^{-1} \ (-5)$
(3)		2CsI	+	$O^{\circ\circ}$	\leftrightarrow	Cs_2O +	I_2	$\Delta G_r^{\circ}(298K) = +17kJ.mol^{-1}$ (+163)

From the free energies estimation, it is likely that the decomposition process taking place is mostly radiolytic and through HO° radical (the free energies estimation at 400K are not significantly modified if we assume that the formation enthalpies and entropies of all compounds are constant from 298 K to 400 K). Moreover, Sulkova [54] highlighted that reactions (1) and (2) take place with no energetic barrier. Considering the forward reactions, reaction (2) is thus the most likely. Another reaction ($2CsI + 2H_2O => Cs_2(OH)_2 + 2HI$) leading to $Cs_2(OH)_2$ gaseous compound has been checked but its free energy at 298 K is close to + 500 kJ.mol⁻¹ which should prevent it to occur in EPICUR facility and also in a NPP containment as temperature is not expected to be over 473 K. It is also consistent with DT4 test that shows no CsI decomposition in humid conditions (RH = 50%) at 105°C.

Even if the humidity effect was not specifically checked with CdI_2 aerosols, if we assume similar decomposition processes for CdI_2 , it leads to reactions (4) to (7). Free energies in brackets refer to Cadmium compounds (reactants and products) considered as solid compounds.

(4) Thermal process: $CdI_2 + 2H_2O \leftrightarrow Cd(OH)_2 + 2HI \quad \Delta G_r^{\circ}(298K) = +192 \, kJ.mol^{-1} (+188)$ (5) Radiolytic processes: $CdI_2 + 2HO^{\circ} \leftrightarrow Cd(OH)_2 + I_2 \quad \Delta G_r^{\circ}(298K) = -318 \, kJ.mol^{-1} (-321)$ (6) $CdI_2 + O^{\circ\circ} \leftrightarrow CdO + I_2 \quad \Delta G_r^{\circ}(298K) = -2 \, kJ.mol^{-1} (-241)$ (7) $CdI_2 + O^{\circ\circ} + H_2O \leftrightarrow Cd(OH)_2 + I_2 \quad \Delta G_r^{\circ}(298K) = -252 \, kJ.mol^{-1} (-256)$ For CdI_2 , the chemical reaction with the lowest free energy is the main chemical path (5). However, it involves the reaction with 2 radicals which is a less likely reaction than reactions (6) or (7) that also have a negative free energy.

From the experimental results on CsI and CdI₂ thermal decomposition (no irradiation) and the estimated free energies for CsI and CdI₂, the thermal process is not likely whereas all the listed radiolytic processes can happen, all leading to the formation of volatile iodine. It is supported by the results in **Fig. 3** to **Fig. 9** (CsI tests), **Fig. 4** (CdI₂ test) and DT4 test, as we did not observe any inorganic release during the thermal phases for both kind of aerosols (CsI and CdI₂).

As a result, the inorganic iodine releases that are observed under irradiation are expected to be mostly from the radiolytic reaction between the aerosols (CsI and CdI₂) and steam radiolysis products (HO^{\circ} for CsI and O^{$\circ\circ$} for CdI₂) through reactions (2), (6) and (7).

2.4 Effect of the deposition of CsI aerosols on Epoxy painted surface

AER3, AER4 and AER5 tests were performed with CsI deposited on Epoxy painted surfaces. The effect of the dose rate can be observed comparing the inorganic release between AER3 and AER4 whereas the effect of the initial CsI aerosols concentration on the paint can be seen comparing AER4 and AER5 tests. Both effects are detailed below.

For these three tests, as soon as gaseous I_2 is released from CsI decomposition, we expect a competition between I_2 adsorption on the paint and I_2 transfer towards the Maypack in the steel loop. A similar kinetics is expected for both phenomena so that they compete with each other. However, as soon as gaseous I_2 is adsorbed on the paint, gaseous I_2 and CH₃I can be released under irradiation as observed earlier [40].

2.4.1 Effect of the dose rate (DR)

As the sources of the facility were changed during the STEM program, we could study the effect of the dose rate on CsI decomposition with AER3 and AER4 whose conditions are close (Table 1). There is a factor 3.5 between the dose rate of AER3 and AER4 tests for which the release is shown on **Fig. 6**. Considering an order one on the dose rate for the iodine release (as usually assumed in

the models), extrapolating the AER3 dose rate to the AER4 dose rate leads to expect a final AER3 inorganic release close to 50% whereas 75 ± 25 % is found (**Fig. 6**) for AER4. Based on the uncertainties on the experimental corrected data (estimated to be $\approx 35\%$ of each data [40]), the dose rate influence can be considered as low/non-significant.

2.4.2 Effect of the initial CsI concentration

The effect of the initial CsI aerosols concentration on the Epoxy paint was checked with AER4 and AER5 tests (Fig. 7). As the concentration was quite low for AER5 (20 times lower than for AER4), the on-line measurements were under the detection limit. However, it was observed that ~ 25 % of iodine had been released from the AER5 coupon during the irradiation (from the post-test activity measurement on the coupon). After having corrected the AER5 raw on-line data, it can be seen on Fig. 7 that the AER5 relative release is \sim 3 times lower than for AER4 at the end of the test which indicates a slower decomposition kinetics. It indicates that the CsI initial concentration has a significant influence on the overall inorganic iodine release. However, another phenomena could also contribute to this observation. As the gaseous inorganic release (from CsI decomposition) can either be adsorbed on the paint, either be transferred towards the maypack, the iodine-paint interaction could have had an influence. Previously, we studied the iodine-paint interaction [40] (interaction between I₂ and Epoxy paint under irradiation). It was found that if the initial iodine concentration on the paint is lower by a factor 10, it leads to an inorganic release lowered by a factor of ~ 2 after about 30 hours of irradiation for initial iodine concentration on the paint ($10^{-4} <$ $[I]_{ads}^{\circ} < 10^{-3} \text{ mol.m}^{-2}$) similar to those of CsI. Basically, it was found that the lower the initial adsorbed I₂ concentration on the paint, the lower the relative inorganic release which is consistent with the observation on Fig. 7.

As the gaseous iodine formed from CsI aerosols radiolytic decomposition can be adsorbed on the paint, it is likely that, during AER5 test, a lower amount of I_2 has been adsorbed on the paint, leading to a lower iodine relative release from the paint than for AER4.

CsI initial concentration is thus expected to have an influence on the release of inorganic iodine. For a modeling approach, this observation indicates that the model of CsI radiolytic decomposition should depend on CsI concentration.

3 Modeling of the data

3.1 Optimization of the radiolytic decomposition model of CsI and CdI₂ aerosols

From AER1, AER2, AER6, AER11 and AER12 tests (quartz and stainless steel coupons), a low amount of organic iodides was detected on the Maypack whereas the inorganic iodine release was found to be significant for all these tests. We assumed that the radiolytic decomposition of CsI and CdI₂ aerosols leads to the formation of gaseous I₂ with the following mechanism and simple model (assumed to be of order one on n(CsI) and the dose rate) (Eq. 1) that was developed in ASTEC-SOPHAEROS severe accident code:

$$CsI_{deposited} \xrightarrow{k_{des}_CsI} 0.5.I_{2(gas)}$$

$$\frac{d n(CsI)_{deposited}}{dt} = -k_{decomposition_aer} \cdot DR \cdot n(CsI)_{deposited}$$
(Eq. 1)

With:

k_{decomposition_aer}: decomposition rate of iodine aerosols (Gy⁻¹)

DR : dose rate (Gy.s⁻¹)

n(CsI)_{deposited}: CsI mole number deposited on a surface (quartz, stainless steel or Epoxy paint) (mol)

A similar equation can be deduced for CdI_2 . The organic iodides formation is assumed to be from the radiolytic reaction between organic pollutions of the gaseous phase (and the ones present in the facility) and gaseous inorganic iodine released from the iodine aerosols as mentioned earlier [55].

This model implicitly assumes that (1) CsI aerosol speciation remains stable during the whole irradiation time (no conversion into another aerosol species like Cs_2I_2) and (2) that CsI aerosol does not chemically interact with the paint. Moreover, as the release mechanism is expected to be heterogeneous, this modeling has to be considered as a first simplified homogeneous approach.

For AER3, AER4 and AER5, the iodine - Epoxy paint interaction model developed earlier [40] was added to the modeling to consider gaseous I₂ and CH₃I adsorption on the paint and release under irradiation.

As the CsI and CdI₂ decomposition model depends on the dose rate, we expect a faster inorganic release for higher dose rates. For each test, a manual fit has been realized to optimise the decomposition rate " $k_{decomposition_aer}$ ".

3.2 Modeling of CsI and CdI₂ aerosols decomposition on inert surfaces (stainless steel and quartz)

On **Fig. 8**, an example of the modeling of CsI decomposition on stainless steel is shown for AER6 test. The organic and inorganic release and modeling are displayed (left), as well as the modeling of CsI decomposition (right). With the appropriate fitted decomposition rate (**Table 4**), the inorganic release evolution is well reproduced as well as the organic iodides (RI) release. The CsI radiolytic decomposition is complete after 30 hours in the AER6 conditions (but could be faster if the dose rate would be higher).

On Fig. 4, the decomposition of CdI_2 into inorganic iodine under irradiation is fast, reaches 80% in 10 hours and is well modelled with the appropriate fitted rate (**Table 4**). Even if the organic iodides release shape is well caught, its amount is overestimated by a factor of 4 at the end of the modeling. It mostly comes from the radiolytic gaseous reaction between volatile organic compounds (VOC) and gaseous I₂ whose modeling is preliminary [5,55].

3.3 Modeling of CsI aerosol decomposition on paint

Fig. 9 shows the modeling of the iodine release and CsI decomposition from AER4 painted coupon. It is observed that at the end of the test, CsI is significantly decomposed (as for the tests performed with stainless steel and quartz coupons at 50 % R.H which is consistent) whereas the remaining iodine amount on the coupon ("Iadspdry" on **Fig. 9**, representing I₂ adsorbed on the paint during the irradiation) could represent up to 25% of the AER4 initial inventory and is assumed to be adsorbed iodine. At the end of the test, adsorbed iodine on the AER4 painted coupon could have been the main iodine contribution on the coupon as CsI contribution represents less that 5% of the initial inventory. Complementary AER4 post-test analyses were made to check how much iodine is washed from the painted coupon (section 1.3). During the washing, CsI is expected to be washed whereas adsorbed I₂ is expected to remain on the paint. The paint rinsing solutions was analyzed and showed that \approx 13 % of the initial iodine inventory was washed. As the global volatilization is 77.5%, it means that \approx 10 % could have been adsorbed on the paint (this could not be verified by a gamma-counting as the coupon washing was performed several months after the test so that the activity of the Iodine tracer was too low to be detected).

Both organic and inorganic species releases are quite well modelled and even if some discrepancies exist, they are reasonable as the uncertainties of the experimental data are close to 30%. In the long term, the discrepancies between the experimental data and the modeling might increase according to **Fig. 9**. The discrepancies might be due to a modification of the CsI speciation on the coupon during the irradiation and/or a chemical interaction of CsI with the paint. Both phenomena are not considered in our modeling approach. They would need to be further evaluated to check their relevancy. If needed, their influence might be eventually introduced in the decomposition kinetics in a second step."

For AER3 and AER5 (**Table 3**), 85% and 75% of the initial iodine inventory remained on the coupon at the end of the test. From the model, the main paint contribution is CsI (70% and 55% respectively). This is explained by a lower dose rate for AER3 than for AER4 and by a lower initial concentration for AER5 (**Table 1**).

The paint washing showed that 43% (AER3) and 40% (AER5) of the initial inventory were washed. The remaining iodine on the paint (assumed to be adsorbed iodine) is thus estimated to be \approx 42% (AER3) and 35% for (AER5) whereas the model predicts \approx 10% for both tests. Even though the global release is well caught by the model, there are discrepancies between the model and the estimated adsorbed iodine on the painted coupon at the end of the test. To ensure that the coupon lixiviation was efficient, Caesium was also quantified in the paint rinsing solution to check if it had been entirely washed by the lixiviation. It turns out that Cs was not entirely washed as 25% (AER3), 12% (AER4) and 37% (AER5) of the Cs inventory remains on the paint coupon despite the lixiviation. The discrepancies between the model and the measurements could come from these phenomena that are not considered in the modeling:

- The CsI speciation that might be modified during the irradiation. Other species might be formed like Cs₂I₂.
- A chemical interaction between CsI (and/or its modified species like Cs°, Cs₂I₂, CsOH...) with the painted coupon through its active functional chemical sites (like amines, amides, alcohol groups).
- As the painted coupon stayed for 12 hours in the rinsing solution (to leave time to CsI to dissolve into Cs⁺ and I⁻), another possibility is the adsorption of iodides ions (I⁻) on the paint in the solution.

3.4 Summary of the decomposition rate optimization

Table 4 shows the optimized decomposition rate for all the tests. All optimized CsI decomposition rates are within the order of magnitude of ~ 4.10^{-5} Gy⁻¹. Based on the uncertainties on each corrected on-line data (35% of each data), the uncertainty on the optimized rates is estimated to be close to 50% (this estimation has been obtained considering the experimental data uncertainties that allowed us to optimize a range for the decomposition rate values and then estimate an uncertainty on this rate). The effect of the parameters that were checked is thus not really significant for CsI. Moreover, CdI₂ has a similar decomposition rate than CsI under irradiation.

It should also be noted that, for all the tests, the amount of iodine predicted with ASTEC to be on the stainless steel vessel surfaces (as adsorbed I_2 or deposited IOx aerosols) is under 4% for all tests. As the corrected on-line data on the Maypack include the steel contribution recovered by the loop and vessel washing, we can conclude that, in the modeling, we expect the small amount of iodine predicted on the steel to have only a very small influence on the estimation of the aerosols decomposition rate in **Table 4** (the relative uncertainty on the decomposition rate is 50%). Basically, if the iodine interaction with steel would not be considered in the model, the estimation of the CsI decomposition rate would not be modified in a significant manner.

Even if the preliminary model of radiolytic reaction between gaseous I_2 and organic pollutions has to be confirmed and refined, there is a reasonably good agreement between the organic iodides data on the charcoal filter and the modeling for all the tests modelled in this paper. It confirms the relevancy to study such reaction that might governs CH_3I volatility in the containment (as shown in previous studies [55,56]) in order to validate a model with more reliable experimental data.

3.5 Discussion

As both CsI and CdI₂ aerosols are soluble, it is expected that an external hydration shell is formed around the aerosols as soon as they are put in contact with the laboratory atmosphere and then when the humidity arrives in the vessel at $80/120^{\circ}$ C. Then, when the irradiation is set up, the water shell is expected to facilitate the access of oxidant radicals (such as HO^o) to CsI or CdI₂ and the formation of volatile iodine. It is expected that both types of aerosols might react in a similar manner to the presence of humidity and irradiation, leading to their complete decomposition in ~ 20-30 hours if the humidity if high enough (> 20 %) and the dose rate close to DR ~ 1 Gy.s⁻¹. In case the iodine aerosols are metallic, multicomponent (made with degraded rods compounds, structure materials and other fission products) and insoluble like in FPT-0 and FPT-1 PHEBUS-FP tests (and partly soluble for FPT-2), the volatile inorganic iodine release is expected to be slower. In fact, if we consider AgI as an average insoluble aerosols, even though the water molecule have access to the external shell of the aerosols, the inner shell are much more difficult to reach. As a consequence, the steam and air oxidant radicals will have a limited access to the deeper shells where most of the iodine might be. It might imply a limited oxidation of these aerosols and a slower decomposition rate than for soluble aerosols (this phenomenon will be investigated in another paper dealing with insoluble aerosols).

In any case, soluble or insoluble aerosols, heterogeneous processes are involved. However, in a first approach, they were modeled with a simple first order homogeneous model from which radiolytic decomposition rates were determined for CsI and CdI₂ aerosols decomposition. Then they have been applied to the decomposition of multicomponent iodine aerosols that settled down on the elliptic floor of PHEBUS-FP tests. The objective was to check if they could increase our understanding of the PHEBUS iodine volatility as they were exposed for several hours/days to the radiation field and could have contributed to increase the iodine concentration in the gaseous phase.

4 Application of the model to PHEBUS-FPT tests - Consequences for the understanding of volatile iodine behaviour in the containment

4.1 Modeling of iodine volatility in PHEBUS-FPT tests with ASTEC-SOPHAEROS V2.1

The development version of ASTEC-SOPHAEROS V2.1 code [5] was used to model the iodine volatility in the containment of PHEBUS-FPT tests, considering a radiolytic decomposition rate for the deposited insoluble multicomponent iodine aerosols on the surfaces. We expect these aerosols to decompose as insoluble compounds like AgI were found to slightly decompose under irradiation in the sump [57] or when it is exposed to oxygen for a long time [58].

The PHEBUS containment nodalisation is the same than the one used in previous papers [59,60]. The objective of this calculation is to check if CsI and CdI₂ radiolytic decomposition rate could contribute to explain the level of gaseous iodine in the PHEBUS containment. Three of the PHEBUS-FP tests were chosen: FPT-0, FPT-1 and FPT-2 [17,18,19] as a significant mass of iodine

aerosols entered the containment and settled down onto the elliptic floor in the bottom part of the containment vessel (Cf. Table 5). In this paper, the modeling results are shown for FPT-1 (similar calculations were also performed for FPT-0 and FPT-2 and lead to similar results and conclusions). The settled mass of iodine aerosols on the elliptic floor has been experimentally quantified at the end of the aerosol phase (\approx 10 hours). For FPT-1, when the washing occurred at \approx 70 hours, different measurements were performed in the sump before and after the washing in order to quantify the iodine mass washed into the sump. However, no measurement was performed on the elliptic floor to quantify the remaining deposited iodine mass right before and right after the washing. As a consequence, we don't know if the iodine aerosol deposited mass on the floor evolved between the end of the aerosol phase (≈ 10 hours) and the washing (≈ 70 hours). Table 5 shows the iodine mass deposited on the PHEBUS FPT-1 floor at \approx 10 hours (490 mg) and the iodine mass washed into the sump at \approx 70 hours (450 mg) (the washing efficiency has not been complete for all the tests and significant mass of iodine aerosols remained on the elliptic floor until the end of the test). From these values, we plotted on Fig. 10 the maximum and minimum iodine masses that could have remained on the elliptic floor right before and right after the washing (≈ 70 hours) and also at the end of the test.

4.2 Discussion of the results

For FPT-1, Fig. 10 shows the remaining mass of iodine aerosols on the floor after having setting up a radiolytic decomposition rate of 4.10^{-5} Gy⁻¹ for the deposited iodine aerosols. The modeling (performed with the current development version of ASTEC-SOPHAEROS V2.1) indicates that, deposited multicomponent iodine aerosols are decomposed in approximately one day. This calculation underestimates in an unrealistic manner the iodine aerosols mass remaining on the floor until the washing occurred (450-490 mg) which provokes in turn, an overestimation of the inorganic iodine gaseous concentration on Fig. 11 (left) by one order of magnitude all over the test. Even if we consider the uncertainties on the estimated masses remaining on the floor (36%) (that would lead to a minimum deposited aerosol iodine mass of 288 mg before the washing), the iodine mass remaining on the floor is still underestimated in a significant manner by the calculation. Moreover, the iodine mass adsorbed on the painted surface is also significantly overestimated all over the test on Fig. 11 (right). This calculation clearly indicates that the radiolytic decomposition rate of soluble CsI aerosols applied to the deposited insoluble multicomponent iodine aerosols of FPT-1 is too high. As mentioned earlier, the insoluble nature of FPT-1 multicomponent aerosols is expected to slow down significantly their radiolytic decomposition rate. For FPT0 and FPT1 (and also for FPT2 for which the aerosols are partly insoluble), we thus expect the insoluble iodine aerosols decomposition rate to be slower than for soluble ones. An estimation of this decomposition rate has

been performed with FPT-1 data (gaseous inorganic iodine concentrations) in order to estimate an upper rate value that would make the FPT-1 gaseous inorganic iodine concentration modeling consistent towards all the other phenomena happening in the gaseous phase and on the containment surfaces. Based on the iodine models that are up to date in ASTEC V2.1 (that include the radiolytic and thermal decomposition models of IOx aerosols whose fate will be treated in a separate paper), and assuming that the multicomponent iodine aerosols are not stable under irradiation, their decomposition rate should not be above $k_{decomposition_aer} \approx 1.10^{-7}$ Gy⁻¹. For FPT-0 and FPT-2, the same conclusions can be drawn. Even though this rate is two orders of magnitude lower than the one for CsI decomposition, its contribution could still play a significant influence on iodine volatility in PHEBUS-FP tests. In order to assess whether the decomposition of these deposited insoluble iodine aerosols could have a significant effect on iodine volatility, the radiolytic stability of an insoluble compound (like AgI) is being checked within the OECD/STEM2 project.

5 Conclusions

The experimental results obtained in the frame of the STEM program have led us to quantify the radiolytic decomposition rate of (soluble) CsI and CdI₂ aerosols deposited on dry surfaces and exposed to gamma irradiation between 80°C and 120°C in presence of 20%-50% relative humidity. Both aerosols completely decompose in 20 to 30 hours for a dose rate close to 1 Gy.s⁻¹. For both aerosols (CsI and CdI₂), the thermal decomposition was determined to be non-significant at these temperatures. The influence of the humidity has been checked. A low humidity (RH = 20%) promotes a higher CsI stability whereas higher RH (50%) promotes CsI decomposition. The dose rate effect has been checked for CsI deposited on painted surfaces and has been found to be low/non-significant. A decrease of the initial CsI concentration leads to a decrease of the inorganic gaseous iodine (and vice versa). The decomposition of CsI on paint has probably led to a significant I₂ adsorption on the paint, limiting its transfer towards the Maypack. CsI and CdI₂ exhibit a similar radiolytic decomposition rate under irradiation.

From the interpretation of the experimental data on soluble iodine aerosol (CsI and CdI₂), a radiolytic decomposition model has been set up in the development version of ASTEC-SOPHAEROS (V2.1) and a decomposition rate has been optimized and applied to the insoluble multicomponent iodine aerosols coming from the primary circuit and deposited on the floor of the PHEBUS-FP tests. It is concluded that the rate of soluble aerosols cannot be set up for insoluble aerosols decomposition as it would overestimates iodine volatility in a non-realistic manner. An estimation of an upper value of insoluble aerosols decomposition rate has thus been performed. We

conclude that the radiolytic decomposition rate of insoluble iodine aerosols should be at least two orders of magnitude slower than the one for (soluble) CsI and CdI₂ aerosols and could still have a significant influence on iodine volatility in the PHEBUS containment despite its low value. Some experiments are planned in the OECD-STEM2 project to check the decomposition rate of (insoluble) AgI on dry surfaces under irradiation. The other phenomena on which uncertainties remain and that were cited in a previous paper [40] are also being checked within different projects. They will be addressed in a separate paper, especially for IOx aerosols behaviour under irradiation. It will help us in refining and quantifying the iodine behaviour in the containment in case of severe accident and the associated Source Term.

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Fig. 1: Simplified view of the experimental EPICUR loop

Test name	Iodine aerosol species	Substrate	Initial Iodine concentration on the coupon (mol(I).m ⁻²)	Average dose rate (Gy.s ⁻¹)	Pressure (absolute bar)	Temperature (°C)	R.H. during irradiation (%)	Gaseous flow in the vessel (L.min ⁻¹)	Studied parameter
AER1		Querta	$(6.1 \pm 0.2).10^{-3}$	0.32 ± 0.03	1.57 ± 0.10	80 ± 2	20 ± 2	0.25 ± 0.01	Reference test
AER2		Quartz	$(3.9 \pm 0.2).10^{-3}$	0.31 ± 0.03	1.67 ± 0.10	80 ± 2	50 ± 5	0.25 ± 0.01	Humidity
AER3			$(6.6 \pm 0.1).10^{-3}$	0.31 ± 0.03	1.58 ± 0.10	80 ± 2	50 ± 5	0.25 ± 0.01	Reference test
AER4	CsI	Paint	$(5.3 \pm 0.3).10^{-3}$	1.08 ± 0.10	1.70 ± 0.10	80 ± 3	50 ± 5	0.25 ± 0.01	Dose rate
AER5			$(2.2 \pm 0.1).10^{-4}$	1.08 ± 0.10	1.70 ± 0.10	80 ± 3	50 ± 5	0.25 ± 0.01	Iodine concentration
AER6		Stainless	$(7.4 \pm 0.2).10^{-5}$	1.03 ± 0.10	1.70 ± 0.10	80 ± 3	50 ± 5	0.25 ± 0.01	Reference test
AER11	steel		$(2.1 \pm 0.1).10^{-4}$	0.97 ± 0.10	3.50 ± 0.20	120 ± 3	50 ± 5	0.25 ± 0.01	Temperature
AER12	CdI ₂	Quartz	$(1.2 \pm 0.1).10^{-3}$	0.86 ± 0.10	1.70 ± 0.10	80 ± 3	50 ± 3	0.25 ± 0.01	Aerosol specie (CdI ₂)
DT4	CsI	Stainless steel	$(9.1 \pm 0.3).10^{-4}$	0	1	105 ± 3	0	0.19 ± 0.01	Temperature and humidity

Table 1: Experimental conditions of the CsI and CdI2 aerosol decomposition tests of STEM project

Test	Iodine aerosol species	Duration of the pre-irradiation phase (hours) (no irradiation)	Duration of the irradiation phase (hours)	Duration of the post-irradiation phase (hours) (no irradiation)
AER1		0	30 (RH = 20 %)	4 (RH = 20 %) + 1 h (dry air)
AER2	CsI	1 (dry air)	30 (RH = 48 %)	4 (RH = 48 %)
AER3		1 (dry air)	30 (RH = 47 %)	4 (RH = 47 %)
AER4		1 (dry air)	30 (RH = 50 %)	4 (RH = 50 %)
AER5		1 (dry air)	30 (RH = 50 %)	4 (RH = 50 %)
AER6		1 (dry air)	30 (RH = 50 %)	4 (RH = 50 %)
AER11		1 (dry air)	28 (RH = 50 %)	3.5 (RH = 50 %)
AER12	CdI ₂	1h (dry air) + 9h (RH = 50%)	30 (RH = 50 %)	10 (RH = 50 %)
DT4	CsI	70h (RH = 50%)	0	0

Table 2: Duration of the three phases of the CsI and CdI2 aerosols decomposition tests of STEM project



Fig. 2: Example of the raw releases profiles on the Maypack for organic and inorganic iodine (raw on-line measurements) for AER2 test

Test name	Iodine aerosol	Substrate	Studied parameter	Activity Balance	Global volatilization	Final corrected on-line RI	Final corrected on-line I2
	species			(%)	(%)	(%)	(%)
AER1		Quartz	Reference test	95.7 ± 4.4	52.7 ± 5.0	1.6 ± 0.2	51.1 ± 5.0
AER2	CsI		Humidity	77.1 ± 3.3	99.0 ± 5.5	2.8 ± 0.3	96.2 ± 5.4
AER3		Paint Stainless steel	Reference test	93.6 ± 6.5	14.6 ± 0.3	0.4 ± 0.1	14.2 ± 6.6
AER4			Dose rate	85.6 ± 3.6	77.5 ± 5.6	6.0 ± 0.4	71.6 ± 4.6
AER5			Iodine concentration	86.6±6.2	24.9 ± 1.0	2.2 ± 0.2	22.6 ± 6.2
AER6			Reference test	86.8 ± 3.2	99.5 ± 8.0	13.2 ± 3.2	85.8 ± 4.5
AER11			Temperature	67.0 ± 1.3	99.6 ± 1.3	10.2 ± 0.9	89.4 ± 1.6
AER12	CdI ₂	Quartz	Aerosol species	89.1 ± 1.5	97.7±0.3	2.7 ± 0.1	95.9 ± 1.9
DT4	CsI	Stainless steel	Temperature and humidity	100 ± 10	0	/	/

 Table 3: Final corrected on-line measurements of the CsI and CdI2 aerosols decomposition tests (%)

Table 4: summary of the conditions of CsI and CdI₂ aerosol decomposition tests and radiolytic

 decomposition rate optimized with ASTEC-SOPHAEROS

Name of the STEM tests	Deposited aerosol species	Type of coupon	[I] _{ads} (mol.m ⁻²)	Average dose rate (Gy.s ⁻¹)	Studied parameter	k _{decomposition_aer} (Gy ⁻¹) (± 50%)
AER1	CsI	Quartz	6.1.10 ⁻³	0.32	Reference test	$(1.5 \pm 0.8).10^{-5}$
(80°C, 20% RH) AER2 (80°C, 50% RH)		Quartz	3.9.10 ⁻³	0.31	Relative humidity	$(4.0 \pm 2.0).10^{-5}$
AER3		Paint	6.6.10 ⁻³	0.31	Reference test	$(1.0\pm0.5).10^{-5}$
(80°C, 50% RH) AER4 (80°C, 50% RH)		Paint	5.3.10 ⁻³	1.08	Dose rate	$(3.0 \pm 1.5).10^{-5}$
AER5 (80°C, 50% RH)		Paint	2.2.10 ⁻⁴	1.08	Iodine concentration	$(0.5 \pm 0.3).10^{-5}$
AER6 (80°C 50% RH)		Stainless	7.4.10 ⁻⁵	1.03	Reference test	$(4.0 \pm 2.0).10^{-5}$
AER11 (120°C, 50% RH)		steel	2.1.10 ⁻⁴	0.97	Temperature	$(4.0 \pm 2.0).10^{-5}$
AER12 (80°C, 50% RH)	CdI ₂	Quartz	1.2. 10 ⁻³	0.86	Aerosol species	$(4.0 \pm 2.0).10^{-5}$



Fig. 3: Corrected inorganic release on the knit-mesh filter for AER1 (20 % R.H) and AER2 (50% R.H) tests -Decomposition of CsI aerosols deposited on a quartz coupon under irradiation at 80°C



Fig. 4: Corrected inorganic and organic releases on the Maypack (left) and remaining CdI₂ aerosols on the quartz coupon (right) for AER12 test under irradiation (80°C and 50% R.H)



Fig. 5: Corrected inorganic release on the knit-mesh filter for AER6 (80°C) and AER11 (120°C) tests - Decomposition of CsI aerosols deposited on a stainless steel coupon under irradiation at 50% R.H



Fig. 6: Corrected inorganic release on the knit-mesh filter for AER3 ($DR = 0.31 \text{ Gy.s}^{-1}$) and AER4 ($DR = 1.08 \text{ Gy.s}^{-1}$) tests - Decomposition of CsI aerosols deposited on painted coupon under irradiation at 80°C and 50%

R.H



Fig. 7: Corrected inorganic release on the knit-mesh filter for AER4 ([CsI] = $5.3.10^{-3}$ mol.m⁻²) and AER5 ([CsI] = $2.2.10^{-4}$ mol.m⁻²) tests - Decomposition of CsI aerosols deposited on painted coupon under irradiation at 80°C and 50% R.H



Fig. 8: Corrected inorganic and organic releases on the Maypack (left) and remaining CsI aerosols on the stainless steel coupon (right) for AER6 test under irradiation (80°C and 50% R.H)



Fig. 9: Corrected inorganic and organic releases on the Maypack (left) and remaining Iodine on the painted coupon (CsI + I_{2ads}, right) for AER4 test under irradiation (80°C and 50% R.H)

	FPT-0	FPT-1	FPT-2	
Core inventory (mg)	36	1120	1570	
Iodine released from the core to the containment (%)	63.8	63.8	56.4	
Containment inventory (aerosol + gaseous) (mg)	23	715	885	
Iodine mass (aerosols) deposited on the elliptic floor at the end of the aerosol phase (≈ 10 hours) (mg)	12 (± 21%)	490 (± 36%)	595 (± 30%)	
Iodine mass (aerosols) washed into the sump (mg)	7 (± 30%)	450 (± 36%)	115 (± 20%) (46% soluble and 54% insoluble)	
Iodine washing efficiency (%)	60	92	23	



Fig. 10: Mass of iodine multicomponent aerosols on the elliptic floor of the PHEBUS-FPT-1 containment vessel until and after the washing (~ 70 hours), considering the CsI decomposition rate



Fig. 11: Modeling of the gaseous inorganic iodine concentration (left) and iodine mass adsorbed on the paint in the PHEBUS-FPT-1 containment vessel until and after the washing (~ 70 hours), considering the CsI decomposition rate (SEQMP = SEQuential Maypack, OLMP = On-Line MayPack)

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