Trapping measurements of volatile iodine by sand bed and metallic filters

Olivia Leroy, Celine Monsanglant Louvet

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Abstract	A series of experimental tests were performed to measure the efficiency of volatile iodine trapping on sand bed filters and metallic filters. These filtering media are used in French nuclear power plants to mitigate releases in case of severe accident. The results show that molecular iodine is more easily trapped than methyl iodide due to its high chemical affinity with stainless steel or with silver and cesium cations included in the filter medium. For methyl iodide, the trapping efficiency is low.

Footnote Information	

Trapping measurements of volatile iodine by sand bed and metallic filters

O. Leroy[1] · C. Monsanglant-Louvet[1]

Abstract

A series of experimental tests were performed to measure the efficiency of volatile iodine trapping on sand bed filters and metallic filters. These filtering media are used in French nuclear power plants to mitigate releases in case of severe accident. The results show that molecular iodine is more easily trapped than methyl iodide due to its high chemical affinity with stainless steel or with silver and cesium cations included in the filter medium. For methyl iodide, the trapping efficiency is low. AQ1

Keywords Source term · Severe accident · Volatile iodine · Sand bed filter · Metallic filter

Introduction

The main objective of a Containment Venting System (CVS) is to preserve the integrity of the containment in limiting the overpressure resulting from severe accident conditions on a nuclear reactor. This venting procedure has a negative effect which is a potential release of radioactive gases and aerosols into the environment if it is not equipped with a retention device.

Currently, some Nuclear Power Plants (NPP) are equipped with Filtered Containment Venting Systems (FCVS). A complete review of FCVS worldwide was undertaken in 2014 [1] and completed in 2018 [2], these reviews were initiated after the Fukuhisma-Daiichi accident. In France, the decision of implementing sand bed filters on all of the 58 PWRs (pressurized water reactors) was made in 1986 and since the mid-1990s all French NPPs are equipped with a filtered containment venting system made of a metallic prefilter located inside the containment in order to confine most of the radioactivity ahead of the sand bed filter that is located outside of the containment.

The R&D programs concerning FCVS began in France in the eighties [3]. The PITEAS program (1982-1986) was devoted to the sand-bed filtering process to retain particulate radioactive products. The FUCHIA program (1989–1991) validated the thermo-hydraulic and filtering behavior of the filtered venting line (sand bed filter and associated piping, without pre-filtration) in full scale. The effectiveness of a filtration device is defined by the Decontamination Factor (DF). The decontamination factor of a filter is obtained by the measurement of the concentration upstream of the filter, divided by the concentration measured downstream of the same filter. The overall retention efficiency with respect to molecular iodine was measured but detailed analysis of results has shown that I_2 is probably trapped by deposition on the internal surfaces of the pipes (DF ~ 10). Complementary experiments performed in 1990–1991 aimed at assessing the aerosol retention efficiency of the metallic pre-filter located inside the containment [4]. Nevertheless, the filtration efficiency of this pre-filter for I_2 has not been established by those experiments. Potential effects of radiations in the FCVS on iodine deposits stability and especially the effect of air radiolysis products had not been accounted for. However, recent source term R&D programs have shown that iodine oxide particles and gaseous organic iodides may contribute significantly to the outside releases in case of venting [5]. The filtration efficiency of metallic filters and of sand bed filters for these species was not investigated in the past.

Sand bed filters and metallic filters are efficient to trap aerosols but are not initially designed to trap gaseous fission products. Among these gaseous fission products iodine is well known to have a high contribution to radiological consequence at short term after the accident. The main volatile iodine species are constituted of molecular iodine (I_2) and

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organic iodide [6] with a special attention to methyl iodide which is the most volatile.

Thus, in the framework of the MiRE project [7] (Mitigation of outside releases in case of nuclear accident) dealing with Fission Product (FP) behavior (especially iodine and ruthenium), IRSN performed experiments to review and evaluate the potential trapping of I_2 and CH_3I through sand bed filter and metallic filter.

After a short description of the IRSN experimental facilities used for these studies, this paper presents the series of tests performed to characterize the trapping of iodine gaseous species by sand bed filter and metallic filter. In addition, complementary tests were carried out in presence of some additives in the sand filter to evaluate the potential trapping increase resulting from the presence of other potential reactive elements released from the core. A test was also performed to evaluate the stability of the trapping in the medium term (about 1 month to get sufficient activity decrease of iodine isotopes), that is to say the potential release of the molecular iodine trapped in the metallic filter due to the accumulated irradiation conditions in the filter itself along time.

Experimental

The metallic filter used in our tests is a small part of a cartridge similar to those installed on French PWRs [6]. It is a PALL® Corporation product. On French PWRs, it consists of 92 filtering elements ("cartridges") of 800 mm length and 60 mm diameter each, with a total filtration surface of 33 m^2. These filtering elements are constituted from the internal to the external parts by a perforated internal grid; a metallic weaved supporting layer; a first filtering metallic medium (PMF FH 200); a second filtering medium (PFM 40); a metallic weaved protective layer. The metallic filter (MF) used for tests is a cylindrical section (52 mm length representing a geometrical surface of 96 cm^2) of the PALL® cartridge. The filter was equipped with two flanges in order to setup the experimental facility (Fig. 1). For tests performed in the PERSEE experimental line, some pieces (40 mm diameter) of the filter was cut and used. Some steel wood was also tested (simulant of MF) to see any difference.

The sand used for tests was mainly a SIBELCO® product from Hostun in Isere Valley, (France). It was sieved between 0.5 and 0.7 mm in order to be representative of sand present on French NPP's with no pre-treatment (SB). To complete the tests, sifted sand is used (SS). Another sand, Fontainebleau type (FS), whose characteristics are perfectly known and reproducible (median diameter 0.25 mm), was used in some tests for comparison. Different sizes of sample were designed for the tests. In severe accident, the operating conditions correspond to a maximum gas velocity of 10 cm/s at the start of the venting procedure. Then the velocity decreases later on when the pressure in the containment decreases. The gas composition, at around 1 bar, is mainly a steam/air mixture with a steam fraction varying between 20 and 30%$_w$.

Experimental molecular iodine test bench

Filtration efficiency facility

The set-up used for measuring decontamination factors of sand bed filters and metallic filters with respect to molecular iodine is schematized in Fig. 2. A gas flow allows the transfer of gaseous molecular iodine through the filter (sand bed

Fig. 1 Section of filter (left)—filter equipped with flanges ready for irradiation (right)
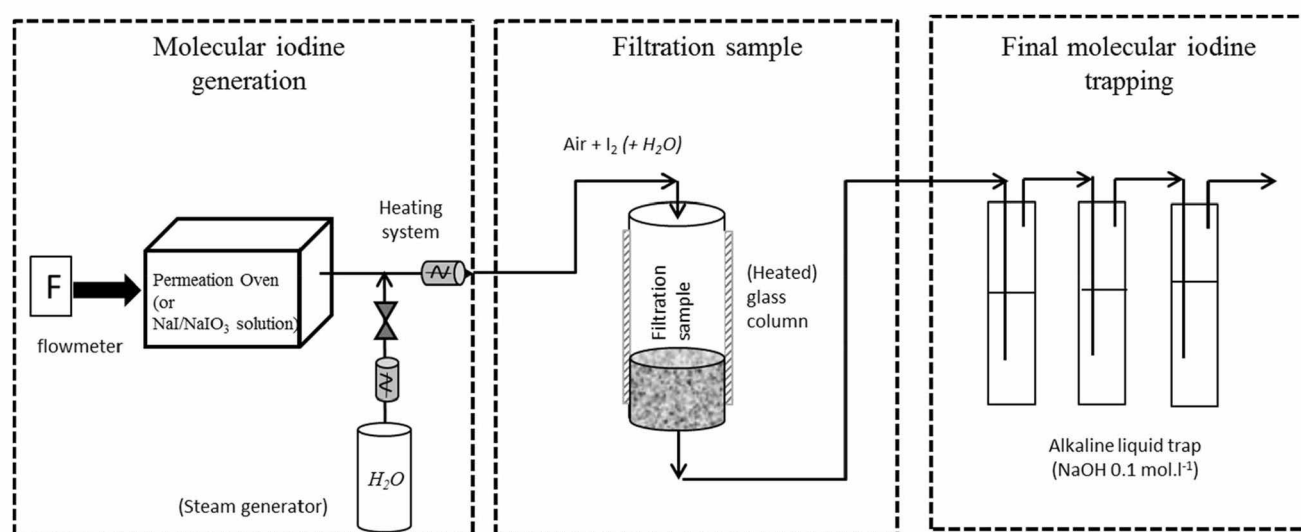
Fig. 2 Experimental facility used for the determination of decontamination factor with respect to molecular iodine

or metallic filter section) where it remains partly trapped. The molecular iodine not trapped by the filter is entrained towards alkaline liquid traps and is dissolved in liquid phase to form iodide and iodate anions.

It can be subdivided into three parts, corresponding respectively to the generation, the filtration sample and the trapping of molecular iodine:

- Two different molecular iodine generation systems are used. For sand bed filter tests, gaseous molecular iodine is produced from an acidic liquid phase based on Dushman's reaction [8] using a sodium iodide and a sodium iodate solution. A glass reactor containing the $NaI/NaIO_3$ solution is heated to 50 °C and it is covered to shelter its content from light. It is instrumented with a temperature sensor (Teflon©-coated PT 100) and a pH probe (Fisher Bioblock® brand, 6 mm gel microelectrode). It is also equipped with a gas inlet, regulated by a flowmeter, at the bottom of the reactor and an outlet for the gaseous reactants on the top plug. The sulfuric acid necessary for the reaction is introduced via a peristatic pump in order to allow a regular production of molecular iodine. At the end of the test, the reaction is stopped by adding a solution of sodium hydroxide, thus the solution reaches a pH higher than 10. For tests performed without steam injection (dry air), a guard flask (not represented in Fig. 2) is added to condense the steam coming from molecular iodine generation. The gaseous I_2 concentration lied between 32 and 426 ppm (as a function of the test), injected upstream of the sand sample.

  For the metallic filter tests, gaseous molecular iodine of 470 ppb is generated using a permeation oven (VICI® 150 °C) with three iodine permeation tubes (VICI®, three

20 cm units) inside. Gaseous iodine is obtained by sublimation of solid molecular iodine pellets. The temperature of the permeation oven is fixed to 100 °C and the argon flow rate is 0.1 L TPN/min (1 atm, 0 °C). This device needs to be calibrated as a function of the operating temperature and pressure. When needed, a steam generator is used in order to fix the relative humidity (**RH**) in the gas flow.

- The filtration module consists of a glass column inside which the sample is placed. The glass makes it possible to limit the retention of iodine on the walls. For the sand bed test, two columns, filled with sand, can be used (Ø 10 cm or 4.6 cm) with a sintered glass type 3 (porosity 16–40 microns) on the bottom to maintain the sand. The metallic filter section is placed in the glass column (Ø 10 cm), heated to 100 °C by heating cords. Thanks to the flanges, the sweeping gas goes radially through the metallic filter from its outside to its inside surfaces.

- The final molecular iodine trapping module consists in three alkaline liquid traps (**NaOH** 0.1 mol/l). At regular time intervals, a sample of trap solution is taken. The liquid samples are then analyzed by inductively coupled plasma-mass spectrometry (ICP-MS) to determine the amount of iodine not retained by the filter, the detection limit is about 8×10^{-9} mol/l.

EPICUR irradiation facility

The experimental set-up used to study the medium term irradiation behavior of molecular iodine previously deposited in the metallic filter consists of an irradiator with ^{60}Co sources (totaling 893 TBq as of September 2012) and a polished stainless steel loop including an irradiation vessel connected

to a selective filter of gaseous iodine species called May-pack (Fig. 3), further details are available in Ref. [9]. The metallic filter previously loaded with iodine is installed in the irradiation vessel. Temperature, pressure and gaseous atmosphere composition are controlled in the irradiation vessel. The specific design allows a homogeneous face velocity of the sweeping gas, radially from the outside to the inside surfaces of the metallic filter. The effect of radiations delivered by the fission products in the metallic pre-filter after an accident is represented by ^{60}Co sources that deliver an average dose rate of several kGy/h. Under the effect of radiation and thermal constraints, the iodine gaseous species released, from the iodine deposited on the metallic filter, are transported to the May-pack device. This system makes it possible to differentiate and measure the various species of iodine released: particulate iodine in a quartz fiber filter (particulate), molecular iodine in a silver clad copper filter called knit-mesh (I$_2$) and then penetrating species (probably organic iodide) in an impregnated charcoal filter. Thanks to the use of ^{131}I to label molecular iodine, continuous measurements are performed on the Maypack filter stages (NaI counters are placed above each filtration stage of the May-pack device) in order to quantify on-line the amount of the different species volatilized and trapped on the selective filters. At the end of the irradiation phase, the iodine filtration system is recovered and the major parts of the experimental loop are dismantled and washed. The different filters, the loaded metallic pre-filter and the washing solutions of the irradiation vessel of the experimental loop are counted after the test in order to compare these post-test measurements with the online measurements and to calculate the overall iodine mass balance at the end. Nevertheless, the online gamma measurements are not as accurate as post-test measurements provided with γ counting on each filter separately after dismantling. The corrective factor is the ratio between the sum of the activities measured on each filter of the stage during the post-test measurements and the activity measured online at the end of the test. So, all the on-line kinetics data are corrected by these ratios to establish the "true" kinetics data presented here.

Description of tests

A series of tests was performed to characterize the efficiency of sand bed filter with respect to molecular iodine. The main studied parameters are: the face velocity, the height of the sand bed and the sand composition (with and without additives). A velocity of 10 cm/s corresponds to the maximum face velocity expected in the sand bed filter during depressurization of the reactor containment following a severe accident. The test matrix is presented in Table 1. For the tests without humidity (SB1 and SB5), the initial moisture content of the sand is about 2% wt. For the tests with wet atmosphere, the humidity of the sand is about 10% wt. In addition to tests performed on sand alone, additional tests have been carried out with additives in order to check the possible iodine retention with these additives which are selected for their affinity for iodine species (like Cs or Ag).

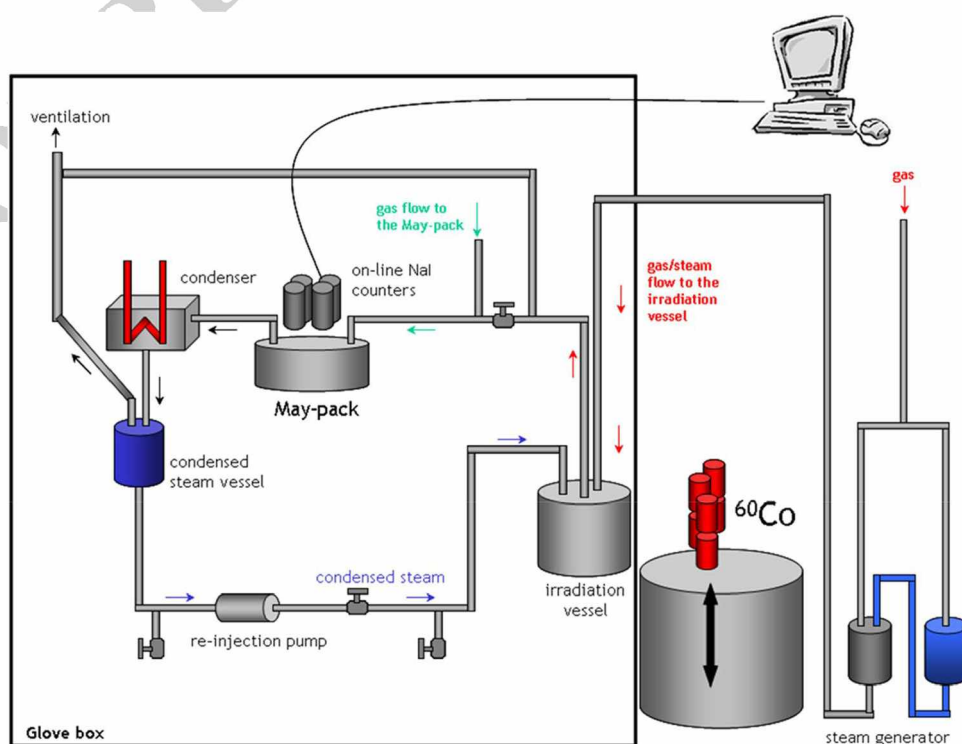**Fig. 3** Process schema of the EPICUR facility

Table 1 Experimental matrix for molecular iodine with sand bed tests

Tests	Parameters tested	Sand—w/wo additive	Column diameter Ø, sand height (cm)	Flow velocity (cm/s)
SB1	Reference—low speed	Dry—w/o additives	Ø 10 cm, 15	0.3
SB2	Humidity	Wet—w/o additives	Ø 10 cm, 15	0.3
SB3	Effect of Cs deposits	Wet—with CsOH	Ø 10 cm, 15	0.3
SB4	Effect of Ag^0 deposits	Wet—with Ag particles	Ø 10 cm, 15	0.3
SB5	High speed	Dry– w/o additives	Ø 4.6 cm, 20	10
SB6 SB6bis	Effect of Ag^+ deposits	Wet—with $AgNO_3$ ($10\%_{/w}$ solution) ($10\%_{/w}$ powder)	Ø 4.6 cm, 20	10

For SB3, a CsOH solution (concentration at 50%w) is deposited at 2.5 cm from the upper part of sand, it corresponds to a target $(Cs/I)_w$ ratio of 2.5 to have an excess of cesium with respect to iodine in proportion which could be encountered in SA conditions. For SB4, a metallic silver powder (2.6 g) is mixed with a fraction of the sand and deposited at the top of the column to reach a target $(Ag/I)_w$ ratio of 8.9. These two additives (Cs and Ag) are present in nuclear accident because cesium is a fission products and silver comes from control rods degradation. The idea was to be assessing possible increase of iodine trapping in more relevant conditions. Another alternative was to check if the initial impregnation of sand by Ag^+ would promote the trapping of iodine. For SB6 and SB6bis, 50 g of Ag (in $AgNO_3$ form) are mixed with 500 g of sand either directly in powder form or in silver nitrate solution and then dried at 60 °C. Tests duration is between 90 min and 180 min depending on the conditions.

The decontamination factor of our experimental metallic filter with respect to molecular iodine (MF_I_2) was investigated. For this test, the I_2 generation kinetics is measured during 1 h using a by-pass line to determine the injected iodine. Then, during 4 h, a flow of I_2 was injected at a face velocity of 1 cm/s through the metal filter maintained at (100 ± 3) °C with a RH of $(40 \pm 4)\%$ ($23\%_w$). Subsequently, the desorption of I_2 from the metal filter was tested under the same conditions but without I_2 flow. The device is heated without air flow up to 100 °C. The time 0 corresponds to the injection of air/steam mixture once the target temperature was reached. The first measurement was performed at 50 min.

Another test, performed on the EPICUR facility (MF_I_2 stab.), was devoted to test the stability of molecular iodine trapped on a metallic filter and then submitted to irradiation. Before the irradiation phase, the metallic filter is loaded with labeled molecular iodine (I^{131}). Molecular iodine was produced in an acidic liquid phase from the Dushman's reaction at 50 °C in presence of radioactive iodine used for labelling (^{131}I, *Perkin Elmer*). After iodine loading and γ counting (13.8 mg of iodine on cylindrical section), the filter is inserted in the irradiation vessel. A specific design allows a homogeneous face velocity of the sweeping gas, radially from the outside surface to the inside surface of the metallic filter. The experimental conditions of the irradiation test, including two desorption phases at 100 °C and 120 °C, are presented in Table 2.

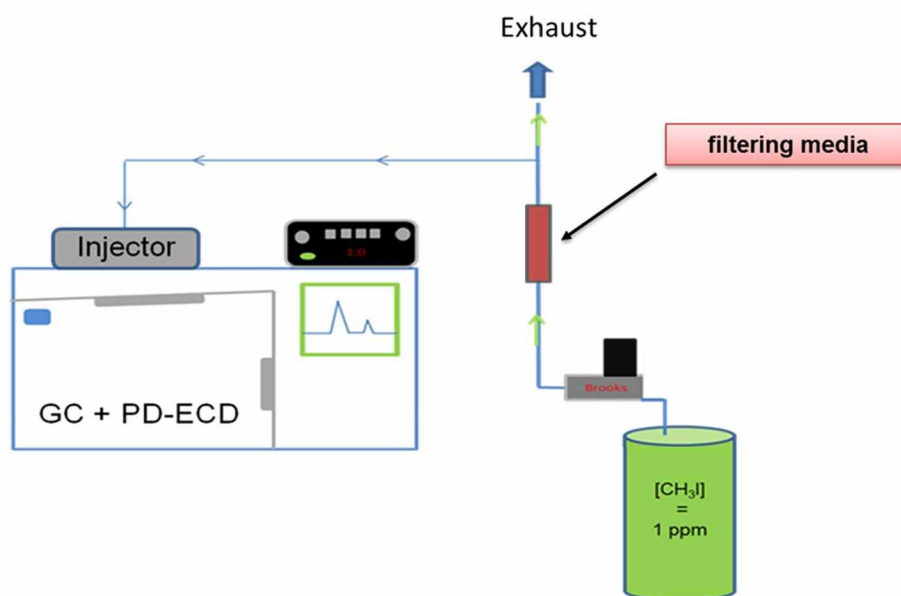# Experimental organic iodine test benches

Filtration efficiency facilities

The test bench (Fig. 4) allows to measure, by gas chromatography (*Perkin Elmer* ® GC), decontamination factors for CH_3I on different filtering media. The detector is a specific pulsed discharge—electron capture detector (PD-ECD) (), very sensitive to halocarbons and other electronegative compounds, and especially to methyl iodide. Organic iodine is obtained from a pressurized bottle at 100 bars and a standard concentration of 1 ppm. The CH_3I concentration of 500 ppb, injected upstream of the sand sample, is prepared by dilution

Table 2 Experimental conditions for the remobilisation test of molecular iodine trapped on metallic filter under irradiation

	Temperature (°C)	Pressure (bas abs)	Gas	Air gas flow rate (g/h)	Water flow rate (g/h)	Dose rate (kGy/h)	Duration (h)
Pre-irradiation phase	100	2.5	Air + steam (35% RH)	51.5	6.8	–	5
Irradiation (phase I)	100	2.5	Air + steam (35% RH)	51.5	6.8	2.0	16
Irradiation (phase II)	120	3.5	Air + steam (35% RH)	48	9.6	2.0	16
Post-irradiation (phase I)	120	3.5	Air + steam (35% RH)	48	9.6	–	5
Post-irradiation (phase II)	120	3.5	Dry air	60	0	–	1

Fig. 4 Experimental facility used for the determination of DF with respect to methyl iodide

with a filtered compressed air flowrate. The gas flowrate at the sand bed filter inlet is 7.7 L/min NTP (Normal Temperature and Pressure conditions: 1 atm, 0 °C), giving an average face velocity of 10 cm/s. The tests were carried out at ambient temperature and relative humidity close to 0%. The filter holder for the sand tests is a cylinder with 4 cm diameter and with 2.5 or 5.0 cm height of sand corresponding to ~ 500 mg for 5 cm.

Additional tests with radioactive iodine (^{131}I) were performed in the PERSEE facility (Fig. 5). It is designed to test filtering media inside sample holders. The test consists in taking two layers of materials. The first layer is the tested filtering media (sand or part of metallic filter in this case). The second layer is a cartridge filled with reference activated charcoal, well-known media with a high decontamination factor for methyl iodide. After conditioning during several hours, both medias are challenged with a pulse of methyl iodine (150 ppb) tagged with radioactive methyl iodine. The measurement of the ^{131}I activity present inside the medias by gamma spectrometry allows determining the DF. The flowrate is controlled by flowmeter at about 18 L/min at NTP.
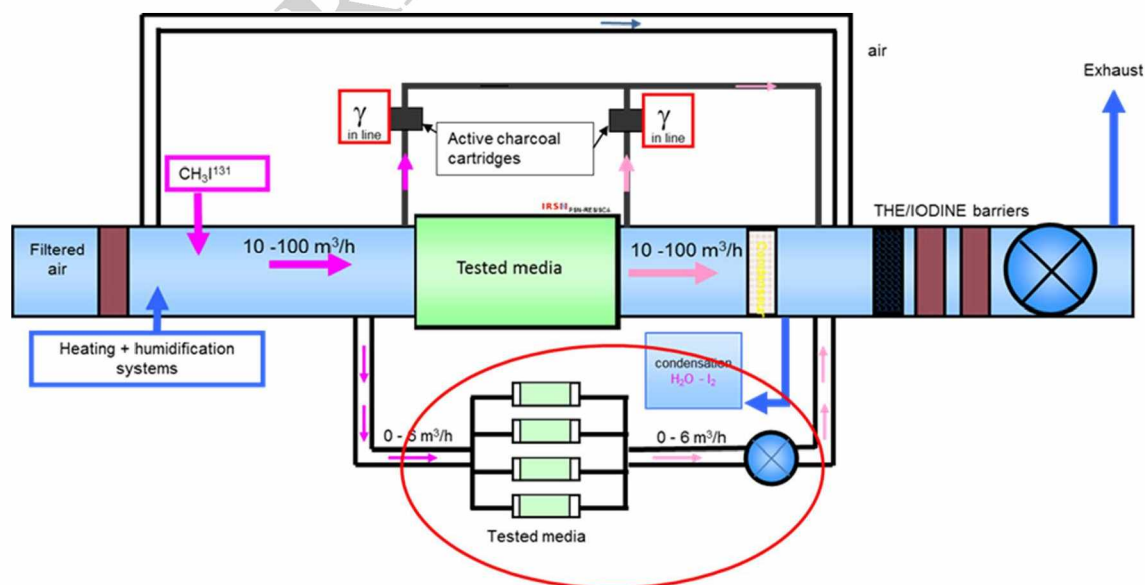

Fig. 5 Experimental facility used for the determination of DF with respect to radioactive methyl iodide

IRMA irradiation facility

In addition to the EPICUR facility, another irradiation facility was used. Indeed, the samples of media to be tested were irradiated in the IRMA cell, also fitted up with ^{60}Co sources (4 sources with a total activity of 896 TBq as of September 10, 2018). This facility allows studying dose or dose rate effects induced by photons on medias or more generally for characterizing the media evolution after irradiation. The number of sources to be used can be adapted according to the target dose rate. It was, as of September 10, 2018, 14 kGy/h at 10 cm of the 4 sources. The duration of the irradiation can vary from a few hours to several weeks depending on the target dose rate and the target integrated total dose. For the present studies, the objects were irradiated until 1 MGy at about 10 kGy/h.

Description of tests

SIBELCO® (SB) and Fontainebleau (SF) sands were tested before and after irradiation (1 MGy), the SF with a small grain size. To complete these tests, different quantities of AgNO$_3$ crystals (between 10 and 30% weight in AgNO$_3$ form) or CsOH crystals (12% or 24% in CsOH weight) were added to the Sibelco® sand. This taggering is carried out either by washing the sand with salt solutions and then by evaporating water in the dark either by mechanical mixing of nitrate powders. Some of these sand samples were also irradiated to check the global chemical stability. All tests are performed at room temperature in air, except the five test performed with labeled iodine performed in temperature and 3%$_w$ in steam as reported in Table 3.

In addition, two tests were carried out on part of a real metallic pre-filter (40 mm diameter) in the same conditions as the last five tests of the Table 3 (^{131}I, 80 °C, 6% R.H., 10 cm/s).

Results and discussion

Molecular iodine test bench

In sand bed filter without additives, the results show (Table 4) that with a low face velocity (0.3 cm/s) or high

Table 3 Experimental matrix for organic iodine and sand bed tests

	Type of material SB: SIBELCO® sand SF: Fontainebleau sand SS: sifted sand	Impregnation	Irradiation
Reference tests	SB	Without impregnation	Irradiated
	SF	Without impregnation	Irradiated
Effect of CsOH	SB	12% CsOH	Non irradiated
	SB	23% CsOH	Non irradiated
	SF	12% CsOH	Non irradiated
	SF	24% CsOH	Non irradiated
Effect of CsOH + irradiation	SB	12% CsOH	Irradiated
	SB	24% CsOH	Irradiated
	SF	24% CsOH	Irradiated
	SF	11% CsOH	Irradiated
Effect of AgNO$_3$	SB	23% AgNO$_3$	Non irradiated
	SF	24% AgNO$_3$	Non irradiated
Effect of AgNO$_3$ + irradiation	SB	11% AgNO$_3$	Irradiated
	SB	23% AgNO$_3$	Irradiated
	SF	11% AgNO$_3$	Irradiated
	SF	23% AgNO$_3$	Irradiated
100% powder	AgNO$_3$ powder	–	Non irradiated
	CsOH powder	–	Non irradiated
"FCVS conditions"	SB (^{131}I test, 80 °C, 6% R.H.)	10%$_{/w}$ AgNO$_3$ in powder form	Non irradiated
	SB (^{131}I test, 80 °C, 6% R.H.)	20%$_{/w}$ AgNO$_3$ in powder form	Non irradiated
	SB (^{131}I test, 80 °C, 6% R.H.)	30%$_{/w}$ AgNO$_3$ in powder form	Non irradiated
	SB (^{131}I test, 80 °C, 6% R.H.)	10%$_{/w}$ AgNO$_3$ in powder form (in layers: AgNO$_3$ first, sand after)	Non irradiated
	SS (^{131}I test, 80 °C, 6% R.H.)	15% AgNO$_3$ (impregnation in solution form)	Non irradiated

Table 4 Decontamination factor for sand bed filter tests with respect to molecular iodine

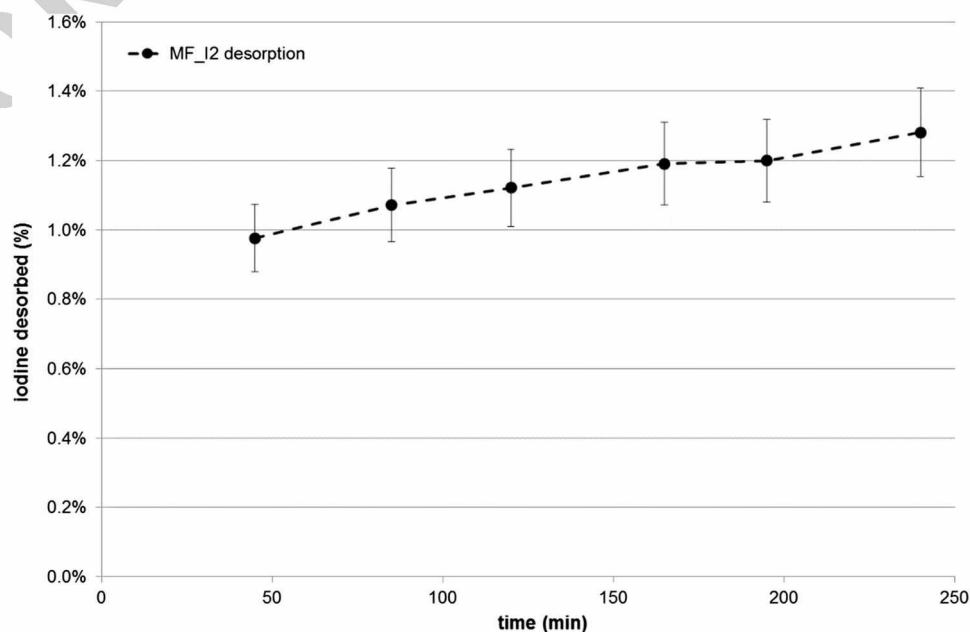| Tests	Parameters	DF
SB1	Reference—low speed	1.0
SB2	Humidity	1.14
SB3	Effect of Cs deposits	173
SB4	Effect of Ag^0 deposits	7
SB5	High speed	0.96
SB6	Effect of Ag^+ *solution*—high speed	7521
SB6bis	Effect of Ag^+ *powder*—high speed	14,080

face velocity (10 cm/s) and with dry or wet sand, the DF is close to 1 (SB1, SB2 and SB5). Less than 1% of molecular iodine is trapped. The sand bed filter alone cannot trap I_2, as expected. The use of additives makes it possible to increase the decontamination factor, even with a high face velocity I_2 is converted into of less or no volatile species. The best results are obtained with the sand doped with 10% of silver (SB6bis) with a DF increase up to 14,000.

Over 4 h, the DF of the metallic filter with respect to molecular iodine is 11. This value may be considered as a low value because nuclear aerosols, composed of fission products and structural materials, already trapped on the filter may react with molecular iodine and so increase the trapping efficiency by chemical interaction [10]. During the desorption phase, at 45 min, a significant desorption of about 1% is measured. It corresponds to the complete phase of heat-up (about 1 h) plus 50 min of tests condition. After the first 45 min, the release rate is relatively slow and constant (0.11%/h). After 4 h, a low but quantifiable

iodine quantity is measured in the liquid traps, about 1.3% (Fig. 6).

Iodine deposition on steel surface has been studied previously [10] but not for filters. The literature is also quite poor at low temperature (~ 100 °C). Usually it is reported that the retention of elemental iodine (I_2) on iron yields metal iodide as FeI_2 [11]. Wren et al. [12] studied, under ambient temperature iodine adsorption and desorption on various stainless steel surfaces (100 h). The adsorbed iodine desorbed partially only under air atmosphere. The major part of iodine didn't desorb indicating that some iodine was irreversibly adsorbed. However, our test duration is too short (4 h) to highlight this stop of desorption which was observed on 100 h duration tests. Nevertheless, it is likely that the same conclusions may apply. So, extrapolating our measured released kinetics leads to a potential maximum release of about 6% at 48 h.

For "MF_I_2 stab" test involving I_2 previously deposited on the metallic filter, results of iodine release are presented in Fig. 7. During the pre-irradiation sweeping phase with moist air (35% RH) at 100 °C, iodine is quickly measurable on the impregnated charcoal. The release is continuing during the irradiation phase of the test. This "organic" release is regularly observed in previous test. It may be attributed to the presence of organic pollution in the loop, in the irradiation vessel or on the filter that may react with gaseous I_2. So this quantity can be re-attributed to "molecular iodine" fraction at the direct outlet of the filter and summed to the knit-mesh activity. A slight increase in kinetics is observed between 100 and 120 °C. We observe also an increase of iodine release after 30 h of irradiation but this increase is probably due to an experimental artefact linked to unwanted

Fig. 6 Evolution of the desorption kinetics (%) at 100 °C and 40% R.H during MF_I2 test
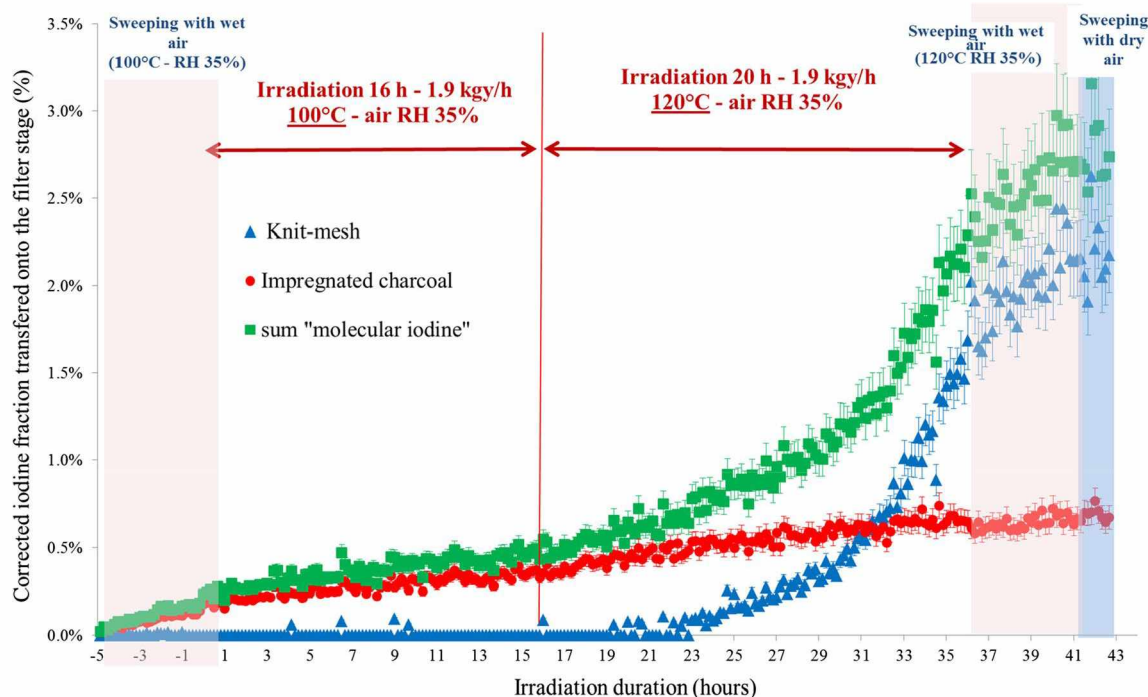
Fig. 7 Iodine fraction released recovered on impregnated charcoal filled circle and knit-mesh filled triangle and the two contribution summed into filled square from the metallic filter during the irradiation test

pressure fluctuations and cumulated temperature effect. The increase of temperature promote the iodine desorption trapped by physisorption process. At the end of the test, the release of iodine measured thanks to the May-pack filter is weak (~ 3% after 36 h of irradiation).

The global release corresponds to the difference between the activity of the metallic pre-filter before and after irradiation, it is expressed in percentage of the activity loaded in the filter before irradiation. The global release is 6.2% of molecular iodine. This result is quite similar to that extrapolated in the desorption test without irradiation. By comparison with pure thermal desorption results and this desorption test under irradiation, it can be stated that there is no significant effect of the irradiation on the desorption process.

Organic iodine test bench

In sand bed filter, as expected, there is no retention of CH_3I. The decontamination factors are close to 1 for all the sand configurations (and materials) tested. Indeed, the sand has a compact structure (no real porosity) in contrast to the materials conventionally used for the adsorption of iodine. Its specific surface area is small (of the order of 1 cm^2/g), to be compared with specific surfaces up to 1000 m^2/g for usual porous materials. In addition, silica is known to be chemically inert. Finally, concerning the tests devoted to improve the filtering efficiency of the sand bed filter with some chemical additives, the results obtained are not conclusive. The treatment of the sand with $AgNO_3$ solution leads only to a slight DF increase (~ 1.4 vs. 1.0) contrary to I_2 (SB6, SB6bis in Table 4). So, the results confirmed that the silver-impregnated sands are not efficient to trap methyl iodide. It is probably due to a not sufficient dispersion of silver species inside the materials as well as the absence of porosity. Indeed, porosity may promote the physical adsorption in a first step of the trapping process before chemical reactions between methyl iodide and silver [13].

For the metallic filter, CH_3I is not trapped. This result is consistent with literature data. Rosenberg [14] investigated the vapor phase deposition of CH_3I on 304 stainless steel plate at 115 °C, in air/steam mixture at 0.35 cm/s and he showed that the deposition velocity was four to five orders of magnitude lower than for I_2. It is worth noticing that the tested conditions are ideal conditions where no aerosol is impacted on the surfaces as it can be in FCVS operating conditions of the metallic filter. Some silver or cesium aerosol deposits should promote the iodine trapping but for methyl iodide this effect should be quite small due to the structure of the molecule, which is a haloalkane. This molecule is not very reactive with respect to surfaces (the reactivity is done via substitutions or binding breaks under high radiation) because it is a saturated compound with a strong C–I binding energy, which is not the case of I_2.

Conclusions

The main conclusion of these test series is that the part of molecular iodine and organic iodine, coming out of the pre-filter stage, is not trapped by the downstream sand bed filter, dry or wet. This is probably due to the absence of internal porosity (like zeolite for instance) which would have promoted the physical adsorption. The addition of silver or cesium inside the sand, leads to a significant increase of the trapping for molecular iodine thanks to the formation of metallic iodide particle; however, it has practically no effect for methyl iodide. The best results for trapping gaseous I_2 are obtained with the sand doped with $10\%_w$ in silver ($AgNO_3$ powder) with a DF increase up to 14,000. Concerning the metallic filter, the trapping is significant (DF ~ 10) for I_2 and it is moreover almost irreversible: less than 5% of trapped I_2 being released after 24 h whereas for methyl iodide, there is no trapping. Research is going to identify porous materials able to trap methyl iodide.

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