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COMBINATION OF OZONE FEED AND WET ELECTROSTATIC PRECIPITATOR: EXPERIMENTAL STUDY OF AN INNOVATIVE SYSTEM TO FILTER GASEOUS IODINE AND IODINE CONTAINING PARTICLES

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Efficient mitigation systems capable of reducing as much as possible the radioactive discharge to the environment in a severe Nuclear Power Plant (NPP) accident are a necessity. Nuclear Power Plants are equipped with filtration systems which have been characterized for aerosol retention in a short term, but to a far lesser extent for the retention of volatile iodine or for the long-term behaviour of the system. After the Fukushima accident, one of the main concerns of the nuclear industry has been to verify the ability of filtration systems to mitigate the possible source term to the environment. Radiotoxic iodine has a significant contribution to a possible source term in a severe NPP accident.

An innovative system has been developed at VTT Technical Research Centre of Finland Ltd. to filter gaseous iodine and iodine containing particles (e.g. I_xO_y*). The system consists of a combination of an ozone feed and a modern wet electrostatic precipitator (WESP). The electrostatic precipitation (ESP) technique is widely used in the industry to filter out impurities (i.e. particles) in gases. The advantage of a WESP is that the impurities are removed from the system with a solution. They can thus directly be transported to a water container, such as the sump in a nuclear power plant. The addition of ozone feed at the inlet of the WESP allows the oxidation of gaseous iodine into particles.*

The efficiency of the system for the filtration of gaseous molecular iodine and methyl iodine has been tested and the subsequent observations are further discussed in this paper. The results showed that the combination of WESP together with ozone feed results in a good filtering efficiency against molecular iodine under air atmosphere and air/steam atmosphere.

I. INTRODUCTION

In the framework of the international PASSAM (ref. [1][2]) programme, an alternative and innovative filtered containment venting system (FCVS) was examined by VTT Technical Research Centre of Finland Ltd. The main objective of this programme was the enhancement of the existing source term mitigation devices and the demonstration of the ability of innovative systems to achieve higher source term attenuation. The innovative technique under study at VTT focused on the combination of an ozone feed and a wet electrostatic precipitator. Since the 20th century, it has been known that corona discharge is able to remove particles from gas streams. Nowadays, many filters using corona discharge exist including Electrostatic Precipitators (ESP), air ionizers and ion wind devices. An ESP removes aerosols from gas flow due to forces induced by strong electric fields (Ref. [3]). The addition of a spray of water mist into the ESP, technique called as Wet ESP (WESP), enhances the particle growth in diameter and the particle charging. Typical filtration efficiency of a WESP for particles is between 99 to 99.9 %.

Incontestably, the WESP technique has shown its ability to filter out particulate impurities in the gas phase, but the technique needs further improvements for the filtration of gaseous impurities. Consequently, the application of WESP under the conditions of a severe nuclear accident establishes a challenge mainly on the filtration of volatile iodine species (including organic iodides). In this studied innovative WESP system an ozone feed is located upstream of the WESP inlet, in order to oxidise the gaseous iodine into iodine oxide particles.

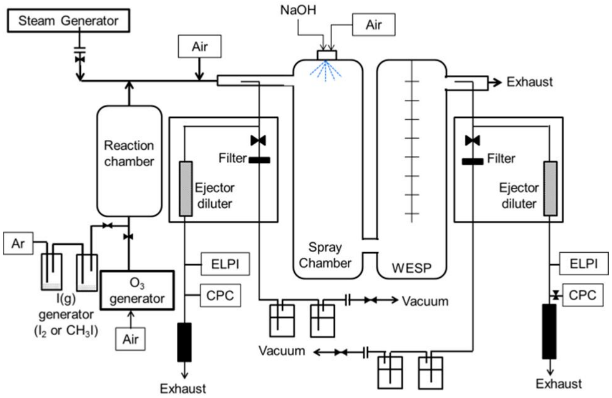
This paper presents the results obtained from testing the efficiency of the WESP in filtering gaseous molecular iodine, methyl iodide and iodine oxide particles at 65°C in steam-containing atmosphere.

II. EXPERIMENTAL

II.A. Facility

The experimental facility consists of a steam generator, a gaseous iodine feeding system, an ozone generator, a reaction chamber (inner diameter: 10 cm, height 68 cm, a droplet spray chamber (inner diameter 21 cm, height 113 cm), the WESP (inner diameter 21 cm, height 113 cm; 4 cm length corona needles, effective length of the central rod electrode 92 cm), and two sampling furnaces (Fig.1). The electrostatic precipitator is of tubular type with small diameter corona needle emitters and a much larger passive electrode comprising of the WESP chamber wall surface.

The filtering of gaseous iodine is performed in four steps. The first step is the oxidation of gaseous iodine into iodine oxide particles (Ref. [4]). Gaseous iodine is produced in a separate section. The iodine vapour generator consists of two plastic flasks filled with coarse particles of solid molecular iodine (ACS reagent ≥ 99.8 %, Sigma Aldrich) or with methyl iodide solution (Stabilized, 99 %, ACROS Organics™). The plastic flasks are held at a constant temperature in a thermostatic bath. The generated iodine vapour is transported out of the flasks by passing the carrier gas through the gas volume above the bed of coarse iodine particles or liquid methyl iodide. Then, gaseous iodine is mixed with ozone. Ozone is generated from the carrier gas air with an additional ozone generator, as well as, ozone is also generated from air in the corona of WESP. As a result, gaseous iodine is converted into iodine oxides (*i.e*., I_4O_9 or I_2O_5). By successive oxidation of I, IO and IO_2 primary products, the final product I_4O_9 is formed with ozone at room temperature. According to Vikis *et al* (Ref. [4]), the molar ratio O_3 consumed per I_2 reacted is 4.5. The maximum ozone concentration generated by the ozone generator is 9000 ppm, thus the used iodine concentration in the experimental facility was less than 2000 ppm.



I Fig. 1. Schematic figure of the experimental facility.

The second step is the growth/agglomeration of the formed iodine oxide particles and the coagulation of particles with sodium hydroxide droplets (dissolved in water). Particulate iodine is dissolved in the droplets, whereas gaseous iodine is absorbed by the droplets. As iodine oxides particles have coagulated with droplets or agglomerated with other particles, the iodine oxide particles grow in diameter and thus the filtration efficiency is also increased.

The third step concerns the iodine chemistry in an electric field of ESP. Corona discharges from the needle-like electrodes are relatively low power electrical discharges that take place at or near atmospheric pressure. The corona is invariably generated by strong electric fields associated with sharp needles. The ions from the corona serve to charge individual particles and droplets as the gas flow carries them through the ESP. Then, these charged particles and droplets drift to the collection electrode due to the electric field.

The last step is the flushing of ESP wall surfaces. At the ESP stage, the collection electrode surfaces of the precipitator are kept clean by flushing with sodium hydroxide solution (doped with sodium thiosulfate $Na_2S_2O_3$ for the experiments with methyl iodide). Hence, there is no re-entrainment of particles to the outlet flow as in case of a dry ESP and the flushing also prevents from the gaseous iodine sorption on the stainless steel wall. The flushing solution also traps the possible gaseous iodine released from the decomposition of iodine oxide particles. The solution is preheated for the experiments performed at 65 °C.

II.B. Chemical analyses and on-line measurements

Gaseous and particulate iodine samples were collected at the inlet and outlet of the WESP (spray chamber + WESP chamber). The scrubber solutions (200 ml) and leachates from filters were analysed with Inductively Coupled Plasma Mass Spectrometry (ICP-MS). All analyses were performed with Thermo Fisher Scientific HR-ICP-MS Element2 apparatus.

Aerosol number size distributions were measured online with Electric Low Pressure Impactors (ELPI) both at the inlet and outlet of the WESP. In ELPI, particles are charged and then collected on 12 different impactor stages according to their aerodynamic diameters and the electrical current on each impactor stage is measured in order to determine the particle number size distribution. ELPI can operate in a particle size range from 7 nm to 10 µm and its time resolution is 1 second. Two TSI (series 3775) Condensation Particle Counters (CPC) were also used to measure the aerosol number concentration both at the inlet and outlet of the WESP. CPCs were able to detect particles higher than 2.5 nm in diameter.

II.C. Matrix of tests

The best parameters related to the WESP performance have been defined previously with tests performed in dry conditions (*i.e.* without steam content) and at room temperature (Ref. [5]). It was concluded that the WESP shows its best filtration efficiency in dry atmosphere with the following conditions in the studied set-up:

- Ozone generator max. output $[O_3]_i$ ~ 9000 ppm;

- NaOH wall flushing: yes;
- Droplet feed (~80 g/m^3, pH~12): yes;
- Max. applied voltage: -25 kV.

For the experiments presented in this paper, the aim was to assess the behaviour of WESP in conditions where steam is present in the carrier gas. Consequently, the facility in general was heated up to 120 °C and the WESP chamber was heated to 65 °C. CPC and ELPI operated at 20 °C and humidity content of the gas flow inside ELPI had to be kept lower than 2 %. Consequently, for the experiments performed with steam, in order to prevent the measurement devices from being damaged, two safety-measures have been executed. At the inlet, the line is heated to 120 °C and thus a second ejector dilutor was added before the ELPI and CPC to decrease the temperature and humidity fraction of the gas flow. However, increasing the dilution ratio after the WESP was not conceivable because of the already low particle number concentration recorded at the WESP outlet. So it was decided to have a maximum temperature of 65 °C inside the WESP in order to condense steam before the outlet sampling. Therefore, regardless of the steam content in the carrier gas entering the WESP, the steam content at the outlet sampling should remain the same.

Two different concentrations of molecular iodine and one concentration of methyl iodide were fed to the facility in the experiments:
- initial $[I_2]_1$[1] ~12 ppm;
- initial $[I_2]_2$ ~100 ppm;
- initial $[CH_3I]_1$ ~6 ppm.

The total flow rate (86 l/min) through the WESP was kept constant, although three volumetric flow rates of steam (100 °C) were tested (11, 34, and 45 l/min) (Table I).

TABLE I. Test matrix.

Experiment number	Steam volumetric flow rates [l/min]	Nature of I(g)
1	11	$[I_2]_1$
2	34	$[I_2]_1$
3	45	$[I_2]_1$
4	11	$[I_2]_2$
5	34	$[I_2]_2$
6	45	$[I_2]_2$
7	11	$[CH_3I]_1$
8	34	$[CH_3I]_1$
9	45	$[CH_3I]_1$

The experiments were repeated in order to have estimation on the reliability of the results.

III. RESULTS AND DISCUSSION

The results are presented as a comparison between the ELPI and CPC measurements at the inlet and at the outlet of the WESP, according to Eq. (1), and defined as the filtration efficiency of the WESP. This efficiency refers to the efficiency for particle number.

$$\text{Efficiency} = 100 - \left[\left(\frac{[\text{aerosol}]_{out}}{[\text{aerosol}]_{in}}\right)\times 100\right] \qquad (1)$$

Similarly, the efficiency related to the iodine mass filtration has been determined from the concentrations measured by ICP-MS at the outlet and inlet of the WESP. Concerning these ICP-MS measurements, 3 results are presented:
- measurements from the liquid trap (*i.e.* total of iodine in gaseous form);
-measurements from the leaching of the filter (*i.e.* total of iodine in particles);
-total amount of iodine (*i.e.* total of iodine gas + particles).
The total amount of iodine is the most pertinent to assess the efficiency of filtering the iodine mass. Indeed, usually the efficiency is lower for the gaseous iodine but the gaseous iodine fraction represents less than 5 % of the total iodine. These efficiencies refer to the efficiency for iodine mass.

TABLE II. Filtration efficiency of WESP for particle number at -25 kV, calculated from ELPI and CPC (*in brackets*) measurements according to the volumetric flow rate of steam fed into the line.

Steam volumetric flow rate [l/min]	$[I_2]_1$ [%]	$[I_2]_2$ [%]	$[CH_3I]_1$ [%]
11	99.73 *(99.58)*	96.75 *(99.89)*	97.1 *(85.5)*
34	99.87 *(99.87)*	95.35 *(99.98)*	95.3 *(85.9)*
45	99.87 *(99.89)*	99.85 *(99.13)*	89.8 *(82.6)*

III.A. Filtration of $[I_2]_1$

The filtration efficiency based on the particle number was higher than 99.5 % (TABLE II). Since the high efficiency was observed for the tested three flow rates of steam, it was difficult to point out the effect of increasing steam content in the carrier gas. The high filtration efficiency was observed for the whole range of particle diameters recorded in the ELPI measurements (Fig. 2). The results obtained from ELPI and CPC

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[1] The concentration corresponds to the initial concentration of gaseous iodine before the dilution. The concentration inside the WESP is consequently much lower.

measurements were relatively similar with a maximum difference of 1 %.

The results from ICP-MS measurements showed also quite high filtration efficiency (> 97.6 %, see Fig. 3) for the total mass of iodine. Moreover, it seemed that the increase of steam fraction in the carrier gas above a certain limit decreased the retention of gaseous iodine in the WESP. At the inlet of the facility, 96 % of iodine mass was in the form of particles and 4 % was in the gaseous form, thus most of the molecular iodine had been transformed into I_xO_y particles.

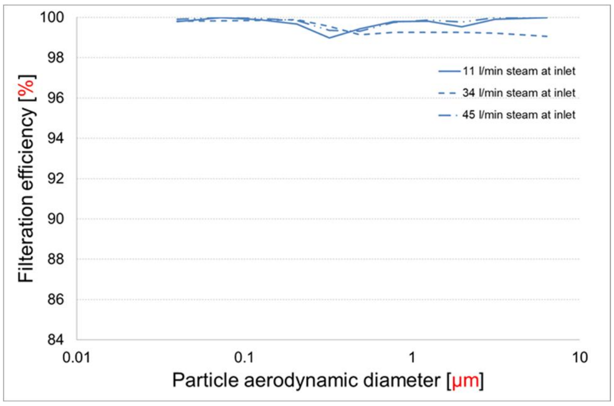

Fig. 2. The filtration efficiency of WESP for particle number at -25 kV, calculated from ELPI measurements according to particle aerodynamic diameter.

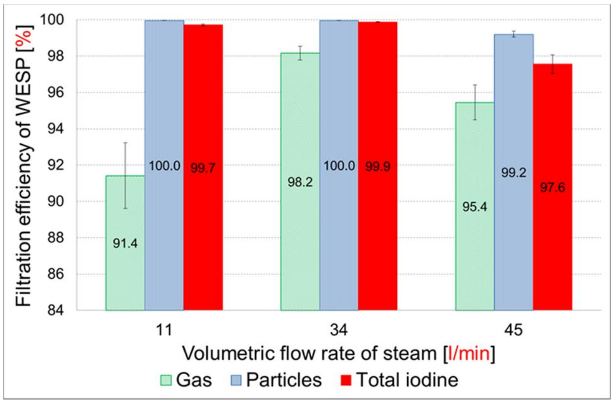

Fig. 3. The filtration efficiency against iodine mass (gas, particle, sum) calculated from the ICP-MS analysis data of particle filter and bubbling bottle samples.

III.B. Filtration of $[I_2]_2$

The filtration efficiency based on the particle number was higher than 95 % (TABLE II). However, it was difficult to point out the effect of increasing steam content in the carrier gas. However, the filtration efficiency was decreased for the particles with a diameter of 0.1 – 0.2 µm (Fig. 4). The results obtained from ELPI and CPC measurements were relatively similar with a maximum difference of 5 %.

The results from ICP-MS measurements showed high filtration efficiency (> 99.4 %, see Fig. 5) for the total mass of iodine. The efficiency was slightly higher than for the lower amount of iodine (see above). It seemed that the increase of steam fraction in the carrier gas increased the retention of gaseous iodine in the WESP continuously. This observation is the opposite of what was observed for the experiments with lower amount of iodine. At the inlet of the facility, 97 % of iodine was in the form of particles and 3 % was in the gaseous form.

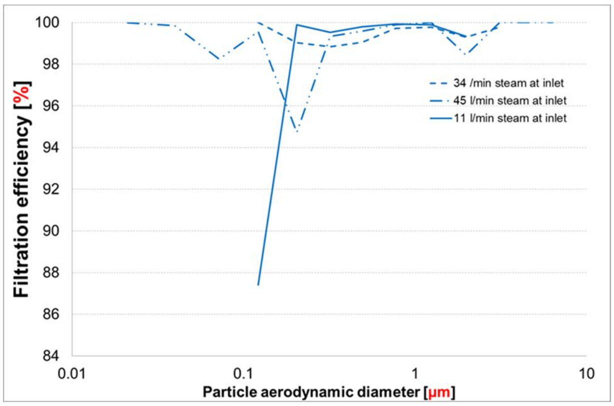

Fig. 4. The filtration efficiency of WESP at -25 kV, calculated for particle number from ELPI measurements according to particle aerodynamic diameter.

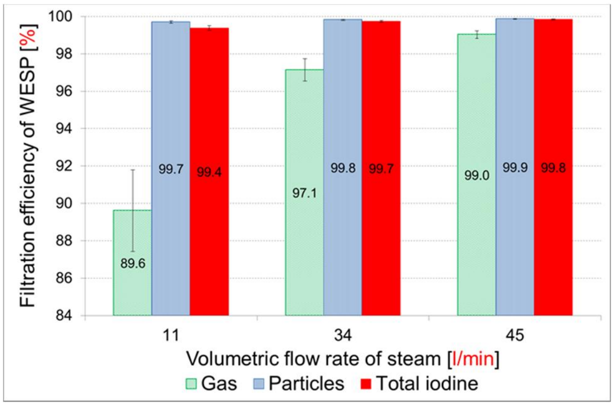

Fig. 5. The filtration efficiency against iodine mass (gas, particle, sum) calculated from the ICP-MS analysis data of particle filter and bubbling bottle samples.

III.C. Filtration of $[CH_3I]_1$

The filtration efficiency against methyl iodide based on the particle number was higher than 82.6 % and 89.8 % by ELPI, which is lower than in the case of molecular iodine (TABLE II). Increasing the steam fraction in the carrier gas tended to decrease the filtration efficiency for the particles. The efficiencies based on the particle number, calculated from the ELPI data, were up to 13 % higher than the ones based on the CPC data. Consequently, CPC measured more particles at the outlet sampling than ELPI. The ELPI measurement uncertainty is very high for the smallest particles less than 40 nm in diameter, which fraction was promoted in the tests with CH_3I in the studied conditions. In this case, CPC measurements may be more reliable.

The highest filtration efficiency was observed for the smaller particle diameters (< 0.3 µm) recorded in the ELPI measurements (Fig. 6).

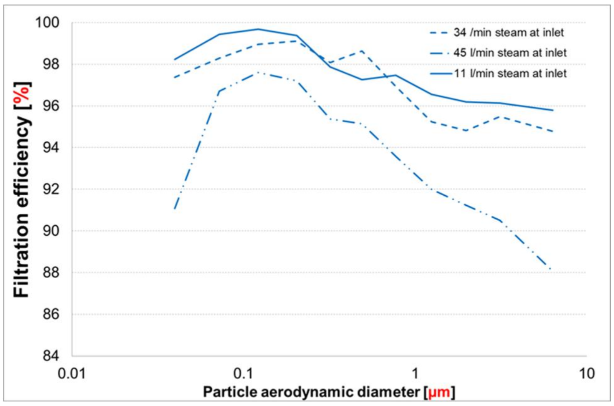

Fig. 6. The filtration efficiency of WESP for particle number at -25 kV, calculated from ELPI measurements according to particle aerodynamic diameter.

The results from ICP-MS measurements showed very low filtration efficiency (> 41 %, see Fig. 7) for the total mass of iodine.

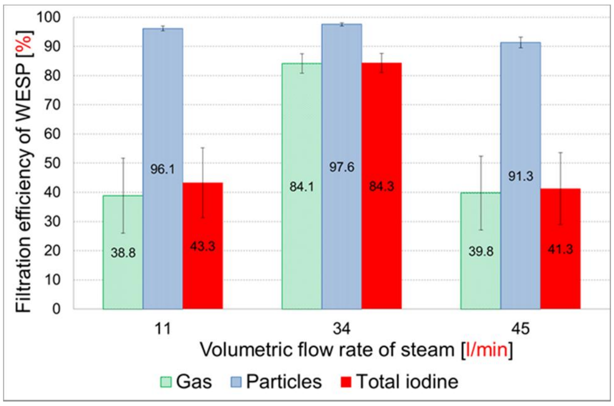

Fig. 7. The filtration efficiency against iodine mass (gas, particle, sum) calculated from the ICP-MS analysis data of particle filter and bubbling bottle samples.

This is explained by the low amount of iodine particles measured at the inlet. At the inlet of the facility, only 5 % of iodine was in the form of particles and 95 % was in the gaseous form. No molecular chemical analysis of the formed particles and the gaseous species has been done. Consequently, it was not possible to determine the reason for the difference on reactivity between the particles formed from methyl iodide and from molecular iodine. The particles measured with ELPI at the inlet sampling of the facility had about the same size (less than 0.1 µm), and the particle mass fraction determined by the ICP-MS analysis of filter leachate was very low

First hypothesis would be that the particles were not formed in the reaction chamber. This would be unlikely to happen, since the residence time before the sampling was 2 minutes and the formation of particles has been already observed from the ozone/methyl iodide reaction in a previous study (Ref. [6]). Another possibility would be that the iodine oxide (I_xO_y) particles decomposed in the presence of steam. In that case, the iodine oxide particles formed between methyl iodide and ozone, and molecular iodine and ozone were different since they present different behaviour. Most probable theory relies on the different particle size. The particles formed from methyl iodide may be smaller, due to the lower vapour pressure, and consequently the particles decompose more easily or the particles need shorter time for the decomposition.

V. CONCLUSIONS

The conclusions from the observed results are valid for the studied range of parameters and for the specific physical size and arrangement of the ozone feed/WESP combination used at the laboratory. The system performance can be adjusted and enhanced by varying e.g. the geometry, flow rate and amount of corona needles. When scaling up the system, the design parameters need to be adjusted according to the desired application.

The work performed in the framework of EU PASSAM project has shown that VTT´s innovation to use “ozone feed and WESP” together enables the filtration of both gaseous molecular iodine and iodine containing particles (*e.g.* I_xO_y). The results obtained for several conditions showed that WESP together with ozone feed trapped efficiently molecular iodine when it was oxidized into iodine oxide particles.

The filtration of methyl iodide was not satisfactory. This was because only 5 % of the measured iodine at the inlet sampling was in the form of particles. It seemed that the formed particles were mainly decomposed due to steam in the gas flow. Further studies will be performed on the location of the iodine oxidation step to avoid particle decomposition before the WESP.

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