Nuclear Engineering and Design 327 (2018) 7–21

Contents lists available at ScienceDirect

Nuclear Engineering and Design

journal homepage: www.elsevier.com/locate/nucengdes

Development and qualification of an innovative wet electrostatic precipitator in view of gaseous iodine filtration on laboratory-scale

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ARTICLE INFO

Keywords:
Iodine
WESP
Nuclear safety
Severe accident mitigation systems
PASSAM

ABSTRACT

An innovative method to filter gaseous iodine based on a wet electrostatic precipitator (WESP) was developed and tested. It is characterized by an ozone feed located before the WESP inlet to oxidize gaseous iodine into iodine oxide particles. Tests were carried out with titanium dioxide particles, first, and then with gaseous molecular iodine (I_2) and methyl iodide (CH_3I). The applied electric voltage between the electrodes, the total flow rate inside the WESP, and the injection of water droplets before the WESP were varied. The filtration efficiency based on the number of particles was calculated from ELPI and CPC measurements at the inlet and the outlet of the WESP. To determine the mass filtration efficiency for iodine, ICP-MS analyses were performed. Once the operation parameters were optimised for the tested conditions, the WESP ensured a mass filtration efficiency of 99.9% and a particle number based filtration efficiency up to 99.9% against molecular iodine. The efficiency for the filtration of methyl iodide was not as high. The filtration efficiency data are presented covering the particle size range of 0.04–2 µm.

1. Introduction

During a severe Nuclear Power Plant (NPP) accident, iodine in a gaseous form, such as molecular iodine (I_2) and methyl iodide (CH_3I), might reach the containment building. To reduce the radioactive discharge to the environment as much as possible, it is essential that the most efficient mitigation systems are installed. Nowadays, the NPPs are equipped with filtration systems that are characterized for aerosol retention for the short term. However, the retention of volatile iodine and the long-term behaviour of the system are less studied. Furthermore, according to the updated fission products chemical behaviour models, iodine oxide particles seem to be one of the major iodine species in the containment. The diameter of newly-born and persistent airborne iodine oxide particles (close to 0.1 µm) seems to coincide with the diameter for which the filtration efficiency of containment venting systems (e.g. sand filter) is the lowest (Raimond et al., 2013).

Within the framework of the international Passive and Active Systems on Severe Accident source term Mitigation (PASSAM) (Albiol et al., 2015) programme, an alternative and innovative filtered containment venting system (FCVS) based on a wet electrostatic precipitator (WESP) system was examined by VTT Technical Research Centre of Finland Ltd. The WESP was investigated to mitigate the iodine source term in gas and particle forms.

Electrostatic precipitator (ESP) removes aerosols from gas flow by means of forces induced by strong electric fields. A high electric field forms a corona on the tip of the centre electrode. A corona is an area of ionized gas that can be recognized by a bluish glow. Ions travelling to other electrodes collide with particles (and droplets) and give them an electronic charge. Particles are charged either before or on the precipitation stage (Riehle, 1997). Charged particles drift to the collection electrode due to the high electric field. The corona discharge process has been well explained by Chang et al. (1991). Electrostatic precipitators are commonly used in industry to remove fly ash from boilers and incinerators (Arrondel et al., 2012; Kim and Lee, 1999), and in the metallurgical industry to control sulfuric acid and particles (Staehle et al., 2003).

The ESPs have many advantages. For instance, they present high filtration efficiency, and the ESPs can handle large gas volumes (Jaworek et al., 2007). The ESPs can be adjusted to handle corrosive materials (Chang, 2003; Chang et al., 2011) and higher operating temperatures (Rinard et al., 1987).

In recent decades, many modifications to the ESP system have been proposed based on the properties of the air to be cleaned. For instance, if the resistivity of particles is too high, they might not be charged high enough for collection. This can be avoided with a wet operation. In a WESP, either the inlet gas flow is humidified or there is a scrubber before the charging/ESP stage (Beltran, 2009; Staehle et al., 2003). Otherwise, the filtration mechanism for particles is similar to ESP. One

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https://doi.org/10.1016/j.nucengdes.2017.12.002
Received 24 August 2017; Received in revised form 30 November 2017; Accepted 1 December 2017

advantage of the WESP is that the water mist in the gas flow condenses on the surface of particles, and thus, particles grow in diameter, which enhances the charging of particles and the subsequent collection of particles in the electric field.

During a severe nuclear power plant accident, a large amount of water vapour released from a reactor cooling circuit can reach the containment building. Therefore, filtration systems must be efficient and selective towards the volatile fission products under the conditions of accident (atmospheres composed of a steam volume fraction of 40–100%). To consider this scenario, the behaviour of the WESP in conditions with various steam factions in the carrier gas was investigated.

On the basis of preliminary tests (Kärkelä et al., 2011) and to meet the requirements specific to the filtration of gaseous iodine and iodine oxide particles, it is proposed in this study to modify the existing (wet) ESP by adding an ozone feed before the WESP inlet to oxidize gaseous iodine into iodine oxide particles. An innovative wet electrostatic precipitator was built and then tested with titanium dioxide particles first. Then, the efficiency of the WESP for the filtration of gaseous molecular iodine and methyl iodide was tested. The design of the WESP and the test set-up, and the subsequent experimental observations are discussed in this paper.

2. Innovative WESP and the test set-up

A photograph and schematic of the experimental set-up used to test the innovative WESP are presented in Figs. 1and 2, respectively. It consisted 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, length of corona needles 4 cm, effective length of the centre electrode rod 92 cm, see Fig. 3), and two sampling furnaces. The experiments were performed in an air atmosphere (“dry conditions”) and in the presence of steam.

2.1. Titanium dioxide aerosol generator

A fluidized bed solids feeder (not displayed in Fig. 2) was used as an aerosol generator. The design was similar to the one presented in Francis et al.’s work (Francis et al., 2010). It consisted of several porous brass nozzles below the bed. Stainless-steel beads, with an approximate 53–150 μm diameter (Surfit™ 316L, Höganäs) were mixed with the TiO_2 powder (pigment Kemira) to aid in breaking apart loose particle aggregates. The average particle number size distribution was determined with SMPS at the WESP inlet. The average Count Median Diameter (CMD) was about 227 nm and the Geometric Standard Deviation (GSD) of particles was 1.58. The electric resistivity of TiO_2 at 25 °C is 10^{10} Ω.m. The line starting from the reaction chamber and upstream was replaced with the aerosol generator in the experiments with TiO_2.

2.2. Generation of gaseous iodine

Iodine vapour was generated by the sublimation of molecular iodine pellets or methyl iodide solution. This process has been used by many authors (Grégoire and Mutelle, 2012; Nicolosi and Baybutt, 1982; Sallach et al., 1986) for molecular iodine I_2 and (Kärkelä et al., 2015) for methyl iodide CH_3I. In other words, the process presents the advantage of a reliable and stable production of vapour and is less dependent on the performer. A gaseous iodine generator was located at the inlet of the experimental set-up. The iodine vapour generator consisted of two gas washing bottles made of Perfluoroalkoxy alkanes (PFA) packed with coarse particles of solid molecular iodine (ACS reagent ≥99.8%, Sigma Aldrich) or liquid methyl iodide (Stabilized, 99%, ACROS Organics™). The bottles were maintained at a constant temperature in a water bath. Iodine vapour was generated by heating the bed of coarse iodine particles or liquid methyl iodide. The generated vapour was transported out of the bottles by a carrier gas. The flexible tubing connecting the iodine generator and the reaction chamber and the valve between them were made of fluoropolymer to prevent any possible attack by the corrosive molecular iodine vapours. These tubings were heated to a temperature higher than the one required for the water bath to avoid vapour condensation.

Argon was used as a carrier gas through the system, and it became saturated with I_2 or CH_3I vapour. When the system was at thermodynamic equilibrium, the molecular iodine partial vapour pressure depended only on the temperature of the water bath. A calculation performed according to the Antoine equation with parameters adopted from Stull’s work (Stull, 1947) expressed this relationship for temperatures below the melting point of I_2 (113.7 °C), and below and above the boiling point of CH_3I (37 °C), (Fig. 4). The conditions used during the tests for gaseous iodine generation (IG) are presented in Table 1.

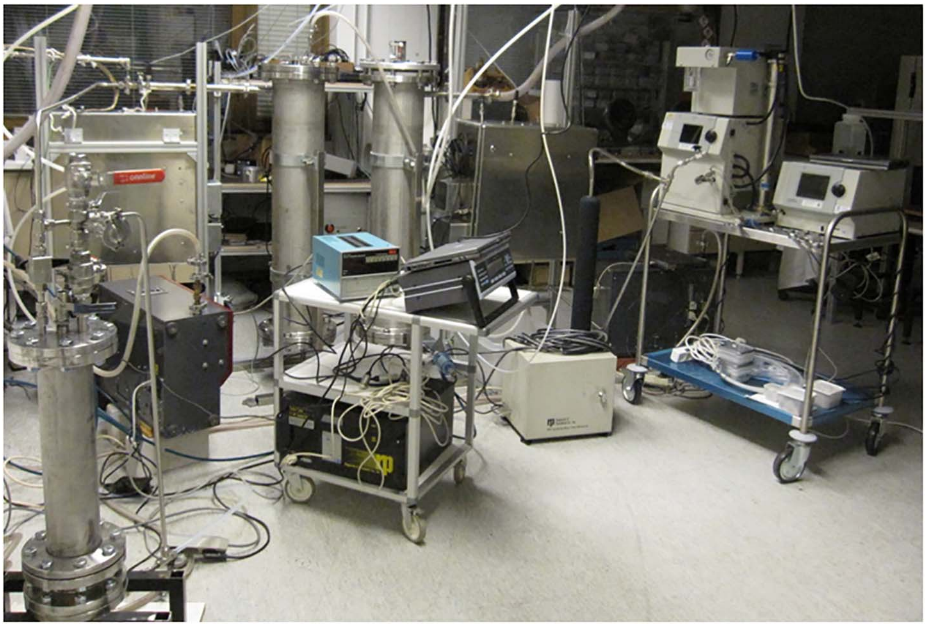

Fig. 1. Photograph of the innovative WESP with online measurement devices at the laboratory.