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Experimental Findings on the Migration of Gaseous Wet I_2 and CH_3I

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1. Introduction

Volatile I_2 and organic iodide are commonly known to move in gaseous or several types of aerosol forms. It was observerd from PHEBUS-FP tests [1] that significant amount of gaseous radioactive iodine (as the forms of I_2 and HI) enters a containment from a RCS without so much sorption in RCS high-temperature tube during fuel degradation [2]. A report on Three Mile Island – II (nuclear power plant accident in 1979, USA) also mentioned that the content of organic iodide in the radioactive emission was more than 40% [3].

A lab-scale set-up including an I_2 (and CH_3I) gas generator, a water droplet generator, and an aerosol collector was installed as a single system with steady control to study how these iodine-related gases and aerosols (or hydrosols in a more specific term in our condition) behave in (the containment of) a nuclear-power plant and further in the environment. The following experiments were planned to look at the effects of water-droplets sizes and radiation field on the migration of volatile I_2 and CH_3I that may occur during an accident in the nuclear-reactor containment.

2. Experimental

The methods to generate volatile iodine species (such as I_2, and CH_3I) that are typically determined in contaminated exhaust air were properly chosen, and water droplets with constant particle sizes were generated and evaporated using the Model 3450 Vibrating Orifice Aerosol Generator (TSI Incorporated, USA). Each experiment was performed at room temperature, and approximate inlet iodine concentration was controlled to be 0.0254 g of solid I_2 or 0.7 mL of liquid CH_3I. The generated volatile gases were introduced into water droplet through cylindrical water jet to look into their behavior.

10 μCi Sodium-22, 1 μCi Cadmium-109, 1 μCi Cesium-137, and 1 μCi Cobalt-57 were put together inside of drying column to see the effect of water-droplet sizes on the migration of volatile I_2 and CH_3I in the presence of radiation field.

To absorb transferred volatile wet I_2, a pure water filled bottle and a 0.1 M sodium hydroxide solution filled bottle were composed into a flow type experimental apparatus. However, volatile CH_3I, unlike I_2, is not absorbed well by a liquid phase such as NaOH solution, so a Tenax tube, which is filled with solid sorbent, was used to adsorb transferred volatile wet CH_3I directly using a pump as shown in Figure 1.

Ultraviolet-visible (UV-vis) spectroscopy and gas chromatography-mass spectroscopy (GC-MS) methods were adopted to analyze the transferred I_2 and CH_3I qualitatively and quantitatively.

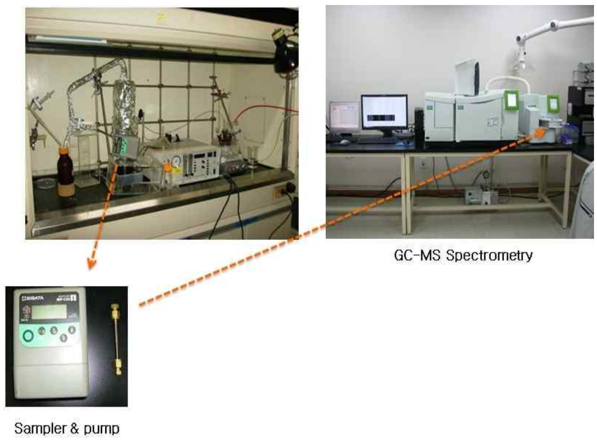

Figure 1: Photo of lab scale set-up and analysis method for a behavioral study of volatile CH_3I from water droplets – air flow.

3. Results and discussion

0, 38, 42, and 48 μm sizes of water droplets could be generated when 0, 80, 60, and 40 kHz frequencies were applied at a nominal operating condition (20 μm orifice diameter, 20 cm^3 syringe capacity, 8.2 x 10^{-4} cm/s syringe pump run speed, and 0.139 cm^3/min liquid feed rate). The formed monodisperse water droplets were well dispersed with 15 x 100 cm^3/min air and diluted 40 L/min air before significant coagulation occurs. Gases of I_2 and CH_3I were generated at about 60°C and room temperature respectively and then led to the cylindrical water jet with the volumetric flowrate of 5cc/min.

The amount of transferred gaseous iodine by adsorption on water droplets was strongly influenced by the existence of water. However, small changes in water-droplet sizes did not influence the transfer of volatile iodine much compared to the case of I_2, but a larger amount of I_2 was transferred with a bigger sized water droplet than a smaller one when the same amount of water was applied. In addition, I_2 concentrations were not detectable without a volumetric flowrate of 5cc/min to introduce the produced I_2 gas to the water line. Similar results were also obtained for volatile wet CH_3I transfer in the case of size changes in the water droplets except the higher concentration of transferred CH_3I gas in the absence of water. CH_3I and H_2O can decompose into various chemical species on exposure to external radiation, so a transferred amount of CH_3I on water droplets were lower under the combination of 10 μCi Na-22, 1 μCi Cd-109, 1 μCi Cs-137, and 1 μCi Co-57 radioactive source discs than the case of absence of the radiation condition as shown in Figure 2. However, a relationship between the transferred volatile CH_3I concentrations and water droplet sizes showed the same trend to the case of absence of the radiation condition. Hydrolysis reaction rates of I_2 and CH_3I with water are very slow in comparison with physical weathering and physical dissolution, and they are only slightly soluble in water. Thus, smaller sized (higher surface areas) water droplets did not help the transfer of the volatile wet I_2 and CH_3I.

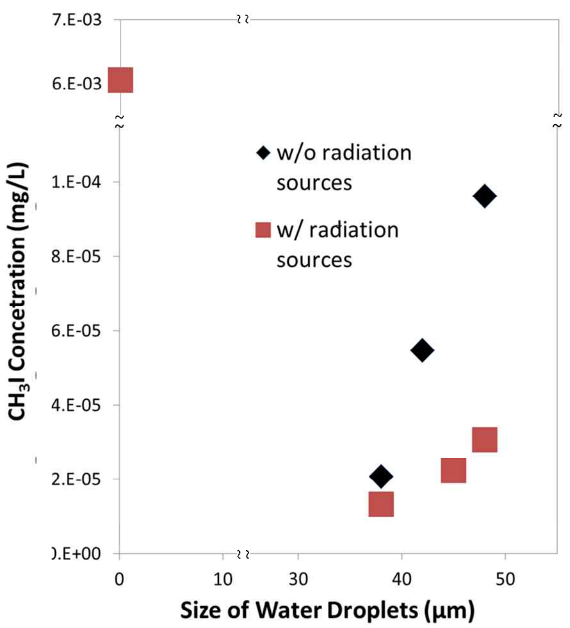

Figure 2: Relationship between transferred volatile CH_3I concentrations and water droplet sizes in the existence of radioactive sources or not.

The results are helpful for an understanding of not only iodine wash-out behavior by water spray but also the I_2 and CH_3I transfer during an SGTR (Steam Generator Tube Rupture). During such an SGTR accident, the I_2 and CH_3I are transferred from the primary coolant (liquid phase) into the second system (which contains the two phases).

Acknowledgements

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