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Behavior of Volatile I_2 and CH_3I from Water Droplets - Air Flow

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1. Introduction

Iodine is a relatively high fission-yield product and volatile nuclide of spent nuclear fuel. It can readily interact with steam and water droplets in a nuclear reactor, and the contaminated aerosols formed can be exhausted in air during emergency venting or after nuclear power plant damage [1]. The released aerosols can further interact both homogeneously and heterogeneously with tropospheric particulate matter, such as smoke, sea salt, or cloud droplets.

However, volatile iodine and organic iodide are seldom discussed owing to a lack of knowledge and experimental data. They are commonly known to move in gaseous or several types of aerosol forms. Therefore, to study how iodine-related aerosols are formed and behave in the containment of a nuclear power plant and further in the environment, lab-scale set-up including an I_2 (or CH_3I) gas generator, a water droplet generator, and an aerosol collector or a sorbent testing tube was installed as a single system with a steady control.

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) [2]. Each experiment was performed at room temperature, and approximate inlet iodine concentration was controlled to be 0.0254 g (1.0×10^{-4} mol) of solid I_2 or 0.7 mL (1.1×10^{-2} mol) of liquid CH_3I. The generated volatile gases were introduced into water droplet through cylindrical water jet to look into their behavior.

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 as shown in Figure 1. 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.

Figure 1: Photo of lab scale set-up for a behavioral study of volatile I_2 from water droplets - air flow.

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.

3. Results and discussion

"The vibrating orifice monodisperse aerosol generator is based on the instability and break-up of a cylindrical liquid jet. A cylindrical liquid jet broke up into droplets by mechanical disturbances. When these mechanical disturbances were generated at a constant frequency and with sufficient amplitude on a liquid jet of constant velocity, the jet broke up into equal sized droplets [3]." So, 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, 8.2×10^{-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 15×100 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 4°C respectively and then led to the cylindrical water jet with the volumetric flowrate of 5cc/min (Figure 2).

Figure 2: Monodisperse (48 μm size) water droplets produced by an aerosol generator (left photo) and volatile I_2 produced from solid I_2 by temperature controlling and air flowing (right photo).

The concentrations of I^-, I_2 I_3^-, and CH_3I in an aqueous phase can be calculated from the absorbancies at 226, 460, 288 (or 350), and 250 nm of UV-vis spectra respectively. Based on the following equations,

$$I_2 + 2OH^- \rightarrow I^- + IO^- + H_2O \quad (k=30)$$

$$3IO^- \rightarrow 2I^- + IO_3^- \quad (k=10^{20})$$

the concentrations of transferred volatile wet I_2 can be calculated from the absorbance of I^- at 226 nm when 0.1 M NaOH solution is an absorbing liquid.

$$(6NaOH + 3I_2 \rightarrow 5NaI + 5NaIO_3 + 3H_2O)$$

As shown in Figure 3, the amount of transferred iodine was strongly influenced by existence of water. However, small changes in water-droplet sizes did not influence so much compared to the previous one, but larger amount of I_2 was transferred with a bigger size of water droplet than smaller one in the same amount of water was applied. Similar results were also obtained for CH_3I transfer in case of size changes in water droplets. Hydrolysis reaction rates of I_2 and CH_3I with water are very slow in comparison with the physical weathering and then physical dissolution, and they are only slightly soluble in water. So, smaller sizes (higher surface areas) of water droplets did not help the transfer of the volatile wet I_2 and CH_3I.

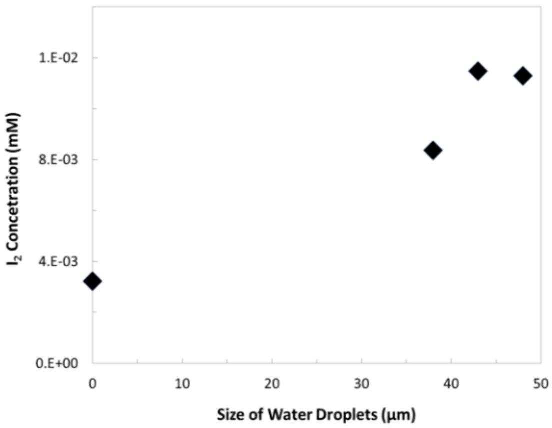

Figure 3: Relationship between transferred volatile I_2 concentrations and water droplet sizes.

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