Effect of water-droplet sizes and radiation-field exposure on the migration of gaseous I_2 and CH_3I adsorbed to water droplets

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Experimental findings on the migration of gaseous I_2 and CH_3I on water droplets of different sizes with or without exposure to radiation field are reported using two different analysis methods: a quantitative analysis based on ultraviolet-visible absorption spectroscopy (UV-vis) and gas chromatography-mass spectroscopy (GC-MS, Clarus 680/ATD-TurboMatrix, PerkinElmer). To study how volatile wet iodine (especially I_2 and CH_3I) behave in the containment of a nuclear power plant and further in the environment during a severe accident of a nuclear power plant, 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. 0, 38, 42, and 48 μm sizes of water droplets could be generated when 0, 80, 60, and 40 kHz frequencies were applied using a Model 3450 Vibrating Orifice Aerosol Generator (TSI Incorporated, USA) 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 a significant coagulation occurs. Gases of I_2 and CH_3I were generated at about 60°C and 4°C respectively, and then led to a cylindrical water jet with a volumetric flowrate of 5 cc/min. To collect 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, and for CH_3I, a Tenax tube, which is filled with a solid sorbent, was used to adsorb transferred volatile wet CH_3I directly using a mini pump.

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. 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.

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).

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