Capture of Organic Iodides from Vessel Off-Gas Streams

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ABSTRACT:

Several gaseous radionuclides, produced during used nuclear fuel (UNF) reprocessing, are present in the off-gas streams of UNF reprocessing facilities. For instance, the dissolver off-gas (DOG) contains a mixture of ^{3}H (as tritiated water), ^{129}I (as $^{129}I_2$ and CH_3I), ^{14}C (as $^{14}CO_2$), ^{85}Kr, and ^{135}Xe. The DOG contains roughly 98% of the total iodine in UNF, while the remaining 2% of total iodine is found in the vessel off-gas (VOG) stream in a mixture of elemental (I_2) and organic forms (CH_3I, etc.). In total, a reprocessing plant needs to capture >99.9% of all iodine to meet federal regulations on plant emissions. Therefore, it is necessary to capture iodine from both the DOG and VOG streams.

Capture of iodine from VOG is complicated by a number of factors including: (i) high flow-rates (roughly 10-100× higher than in DOG), (ii) low concentrations (between 5-100 ppb compared to ppm levels in DOG), and (iii) presence of volatile organic species from solvent extraction. These conditions create a very dilute system where the iodine present in the off-gas stream is made up of a complex mixture of organic iodides ranging from methyl iodide (CH_3I) to iodo-dodecane ($C_{12}H_{25}I$). To efficiently capture the organic iodides from VOG, studies must be carried out experimentally and analytically to determine an effective adsorbent and understand the adsorption properties.

Silver adsorbents have long been considered for iodine adsorption due to the high strength of the Ag-I chemical bond, which helps to prevent desorption of iodine after capture. Additionally, studies have shown that adsorbents impregnated with reduced-silver (Ag^0) can achieve higher decontamination factors (DFs) and iodine loading than their unreduced (Ag^+) counterparts. Therefore, this study proposes to first compare the adsorption capacity of three Ag-containing adsorbents: reduced Ag mordenite (Ag^0Z), reduced Ag-functionalized aerogel (Ag^0-aerogel) and Ag alumina. With the selected adsorbent, adsorption experiments and material characterizations will be performed to determine the adsorption kinetics and reaction pathways for the organic iodides. In addition, the oxidation or "aging" of the reduced-Ag adsorbents in streams containing H_2O and NO_X will also be investigated.

To be able to design the necessary capture system for this off-gas stream, we need to develop adsorption models that account for the various reaction pathways between organic iodides with silver-containing adsorbents and the aging of the silver adsorbents, in addition to the off-gas stream temperatures, compositions, and flow-rates that are expected to occur under realistic plant conditions. Therefore, this study also proposes to develop adsorption models to simulate iodine uptake by reduced-silver adsorbents. These models will need to be flexible and multifaceted such that they can account for the effects of temperature, gas composition, gas flow rates, mass transfer, and all the relevant reaction mechanisms between organic iodide species and the reduced silver sites. Furthermore, these models will need to account for the "aging" of the materials caused by oxidation of the reduced-silver sites.