Mechanistic Understanding of Silver Sorbent Aging Processes in Off-Gas Treatment

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ABSTRACT:

This effort will address a major problem in the treatment of nuclear-fuel reprocessing off-gas steams – the loss of capacity of adsorbents due to aging processes. In particular, the capacity for iodine removal by columns of silver-exchanged mordenite (Ag^0Z) and silver-functionalized silica aerogel (Ag^0-aerogel) are significantly decreased over extended periods of exposure to gas streams containing air, H_2O, and NOx. We will collect experimental data on sorbent deactivation and will employ that information to extend models we have developed for capture of gases in adsorption columns, incorporating the effects of aging and deactivation.

In order to develop transport models that can predict the adsorption capacity over long periods of time, descriptive models for adsorption must also account for the aging processes. Towards this objective, an experimental and modeling program is proposed to obtain data and develop models to describe the deactivation process of the adsorbents when exposed to off-gas species. Building upon recent experimentation at national laboratories, experiments of aging with H_2O, NO, and NO_2 in air over a range of operating conditions will be conducted on Ag^0Z, Ag^0-aerogels, and Ag-coated glass beads for aging times of 1 to 6 months. Approximately 75 aging experiments are planned. These aged adsorbents will be analyzed for chemical and structural changes using several characterization techniques including scanning and transmission electron microscopy (SEM and TEM) for surface structural changes, BET (Brunauer, Emmett, and Teller) analysis for surface area and porosity changes, mercury porosimetry for pore distribution changes, X-ray diffraction (XRD) and nuclear magnetic resonance (NMR) to determine the amount of reduced silver converted to silver ions, X-ray absorption fine structure (XAFS) and pair density function (PDF) to determine the chemical environment of Ag, and X-ray photoelectron spectroscopy (XPS) for surface elemental analysis to determine amounts of Ag, Ag_2O, $AgNO_2$, $AgNO_3$ and other formed compounds and estimate their impact on aging. These data will support model development for the aging processes that will consider the surface reactions of the adsorbents with the reduced silver. Aging of Ag-coated glass beads will provide additional information on the reactions of these components with reduced silver in the absence of interferences from the structural properties of the adsorbents. To determine the impact of aging on the adsorbent capacity, I_2 adsorption studies will be conducted with aged and fresh adsorbents to quantify the loss of capacity.

These data will be used for model development and validation. Our current two-dimensional models combine equilibrium, mass transfer, and kinetics, and include a detailed description of the microscopic physical adsorption process and coupling with macroscopic transport. The microscopic adsorption model will be extended to incorporate experimentally validated aging mechanisms in order to describe the real behavior of the adsorbent over extended periods of time. Models developed in this study will be made available for more accurate design and performance prediction of engineered off-gas treatment systems.