Radioiodine Speciation Impact on Ag-Activated Carbon Immobilization in Cementitious Environments

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Iodine-129 (^{129}I) is a waste product from civilian and defense nuclear material processing. The almost exclusive process used for ^{129}I removal from liquid and gaseous waste streams involves precipitation onto silver (Ag) amended substrate. The objectives of this study were to evaluate the impact of iodine speciation (I^-, IO_3^-, and org-I) on: 1) uptake by Ag sorbents in a cementitious aqueous environment, and 2) leachability from Ag granular activated carbon (Ag-GAC) embedded in cementitious materials. I^- sorption K_d values ($[I_{solid}]/[I_{liquid}]$) incrementally increased to $>10^5$ L/kg as the percent of Ag on the GAC increased from 1 to 14 wt-%. Concurrent with increases in Ag percent on the GAC, the pH decreased from 12.2 to 11.2 and oxidation/reduction potential (ORP) increased from -60 mV to +40 mV in the cement-simulant/AgGAC suspensions. AgGAC K_d values for IO_3^-, org-I, and I^- were 32, 1212, and >6000 L/kg. All three iodine species had significantly greater AgGAC K_d values under reducing conditions than under benchtop/oxic conditions. Control GAC samples without Ag amendments had little capacity to bind any of three iodine species, with K_d values <10 L/kg. Iodine K-edge XAS analyses of grout containing Ag-iodine-GAC confirmed that little IO_3^- was bound to the Ag-GAC, irrespecitve of whether slag (a chemical reductant) was included ($Grout_{+slag}$) or was not included ($Grout_{-slag}$) in the grout formulation. For the grout containing the I^- amended Ag-GAC, the iodine no longer existed as AgI, instead its speciation had changed. Desorption studies demonstrated that initial iodine speciation influenced the extent of iodine immobilization by the AgGAC and leachability from AgGAC-embedded grout.