DEPARTMENT OF CHEMICAL & ENVIRONMENTAL

PH.D. DISSERTATION DEFENSE

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Release and Control Strategies of Hexavalent Chromium in Drinking Water Distribution Systems

Abstract

The occurrence of hexavalent chromium Cr(VI) as a carcinogen in drinking water is widely reported. Cr can accumulate in corrosion scales in drinking water distribution systems (DWDSs) and act as the potential geogenic source for Cr(VI) release. One Analysis of U.S. EPA nationwide Cr(VI) monitoring database shows that there was a 41% chance for an increase in Cr(VI) concentration in DWDSs, and the increase was strongly correlated with the presence of free chlorine as the residual disinfectant.

The goal of this dissertation is to elucidate the mechanisms of Cr(VI) formation through the oxidation of Cr solids by disinfectant free chlorine, and to provide control strategies for Cr(VI) formation in DWDSs. First, X-Ray Absorption Spectroscopy (XAS) analysis discovered that zerovalent Cr(0) coexisted with trivalent Cr(III) solids in the corrosion scales. Because Cr(0) exhibited a much higher reactivity than Cr(III) in formation of Cr(VI) by chlorine, the oxidation of Cr(0) solid in the iron corrosion scales was the dominant reaction for the Cr(VI) formation in drinking water. Second, the corrosion control strategies through adjustments of chemical water parameters were investigated. The results showed an increase in pH, silicate, alkalinity, and calcium suppressed Cr(VI) formation that was mainly attributed to *in situ* surface precipitation of new Cr(III) solids on the surface of $Cr(0)_{(s)}$, including $Cr(OH)_{3(s)}$, $Cr_2(SiO_3)_{3(s)}$, $CrPO_{4(s)}$, $Cr_2(CO_3)_{3(s)}$ and $Cr_{10}Ca(CO_3)_{16(s)}$. The Cr(III) surface precipitates were much less reactive with chlorine than $Cr(0)_{(s)}$ and suppressed the Cr redox reactivity. Adding phosphate either promoted or inhibited the Cr(VI) formation, depending on the phosphate concentration. Third, it is investigated that the effects of bromide and manganese on Cr(VI) formation through the oxidation of Cr(0) solid by free chlorine. The present of Br^- and Mn^{2+} significantly enhanced the Cr (VI) formation due to their catalytic effects. Br^- and Mn^{2+} can react with free chlorine to form free bromine and manganese dioxide, and they have higher reaction kinetics with Cr(0) than free chlorine. The outcome of this study aimed to provide control strategies to minimize Cr(VI) formation by inhibiting Cr(0) reactivity in drinking water distribution systems.

Dr. Haizhou Liu, Chair

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