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U and Th source term characterisation in selected Au tailings of the Witwatersrand, South Africa: A geochemical modelling and reaction network approach

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In this study an iterative geochemical reaction and reaction network modelling methodology is used to identify the source terms and evaluate the geochemical behaviour of U and Th in Au mine tailings in the Witwatersrand, South Africa. This study forms part of a larger radiological assessment study conducted for specific mines in the Free Sate and Klerksdorp Au mining areas. A number of analytical techniques and methodologies were applied. The analytical methods used were based mostly on South African legislative and best practice requirements, although some, specifically the adsorption tests, needed to be developed and indicates gaps in current legislative requirements regarding the geochemical impact assessments of source term characterisation studies. The laboratory data is used to develop the models. This methodology proved to be a useful tool in understanding the geochemical dynamics of U and Th in Witwatersrand Au tailings dams.

Thorium was shown to be immobile in the tailings impoundment. The modelling indicated that the geochemical behaviour of U is governed by the dissolution of pyrite and subsequently uraninite, U complexation and acid neutralisation reactions, by carbonates added to ore in the plant process. The onset of pyrite oxidation has an autocatalytic effect of subsequent reactions, including uraninite oxidation, U(IV) oxidation to U(VI) and the oxidation of Fe(II) to Fe(III) and precipitation of ferric oxyhydroxides, such as goethite. The autocatalytic effect is either promoted or inhibited by positive and negative feedback loops caused by aqueous speciation and acid neutralisation reactions within the impoundment. Adsorption in Witwatersrand tailings impoundments is shown to be negligible and dissolution and precipitation reactions dominate U and Th geochemical behaviour in the tailings impoundments. U specifically is shown to leach from the base of the tailings at concentrations between 67 and 95 µg/l. In addition, the study indicates that the spatial nature of the various geochemical zones within the tailings impoundments may also have important implications for leaching of not only the radiological, but also the metal and metalloid and anionic contaminants and acidity.