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The effect of competition from other metals on nickel complexation by α -isosaccharinic, gluconic and picolinic acids

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The UK has an underground repository concept with a combination of engineered and natural barriers. Cementitious materials will produce high pH conditions for at least 1 Ma and surfaces for sorption which will greatly reduce the solubility of many radionuclides. Likely to be in the repository are many organic ligands, as inherent components of wastes or due to the degradation of organics, e.g. cellulose. It has been shown that cellulose degradation products, e.g. α -isosaccharinic acid (ISA), can substantially increase the solubility of radionuclides. Picolinates are in some waste-streams due to the use of picolinic acid in decontamination. Gluconic acid is structurally similar to ISA, a strong ligand useful for comparison purposes and may occur in a repository as a cement additive. From pH 11.5 to 13.5, Ni is fairly insoluble. The 3 ligands increase its solubility by forming aqueous complexes - a process that is fairly well understood. The purpose was to investigate the effects on Ni complexation, of competition from other metals. A repository will also contain Fe metal so experiments were performed in its presence and absence. Predictions of Ni solubility were made for each system to aid the interpretation of competitive effects.

Measured [Ni] in the absence of other metals showed good correlation with calculated values. There was evidence for slight sorption of Ni to Th and Eu phases formed. There was no significant effect due to other metals and/or solid phases being present. In general, the calculated [Ni] trend was followed with picolinate. Th and Hf had little effect on Ni solubility in the presence of picolinate, Co reduced [Ni], which might be expected as it complexes in a similar fashion, above pH 13 all metals caused a significant reduction in [Ni]. This may have been due to sorption to solid phases, or coprecipitation, with Ni being scavenged out as the lowest inventory metal.

All the Ni was complexed by gluconate. Eu had no significant effect at the highest Ni concentration. Hf, Co and Th reduced [Ni] across the pH range, probably due to complexation with gluconate. Eu had a marginally greater effect at a [Ni] = 10^{-3} M, suggesting a very slight amount of complexation. Hf again had little effect. The Th results were the most difficult to explain. At [Ni] = 0.01 and 0.0001 M, Th affected [Ni] as though forming strong gluconate complexes, but not at 0.001 M. At the lowest [Ni], all competing metals significantly reduced [Ni]. Th had complexed strongly with gluconate, causing Ni to precipitate. Hf had a similar effect to. Co significantly reduced [Ni] in line with its predicted effect. Eu had a significant effect on [Ni] reducing it from the predicted 10^{-4} M to ca. 2×10^{-6} M.

Fe was predicted to have no effect on [Ni] by complexation with ISA. The measured [Ni] was lower than the calculated in virtually all cases, including those with no competing metal. This may have been caused by sorption to Fe(s). No significant competition effects from other metals were observed.

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