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Behaviour of radionuclides during microbially-induced mining of nickel at Talvivaara, Eastern Finland

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ABSTRACT

The Talvivaara mine in Eastern Finland utilizes microbe-induced heap leaching to recover nickel and other valuable metals (Zn, Cu, Co) from a black schist ore. In addition to the target metals, the ore contains uranium at a concentration of 17 mg/kg, incorporated as uraninite (UO_2). Uranium oxidizes from the U(IV) to U(VI) state during leaching and dissolves as the uranyl ion (UO_2^{2+}) in the acidic pregnant leach solution. Mobilisation of uranium has caused sufficient concern that plans have been developed for uranium recovery. The aim of this study is to generate new data leading to a better understanding of the fate of its radiotoxic daughter nuclides, primarily ^{226}Ra, ^{210}Pb and ^{210}Po, in the mining process.

It is shown that uranium daughters mostly remain in the heaps during the leaching process and are associated with secondary minerals, including jarosite, goethite and gypsum. Thorium and progeny (^{232}Th plus ^{228}Th, ^{228}Ra) are also mainly retained. High sulphate concentrations in the acidic solutions limit the solubility of radium by incorporation in the crystal lattices of precipitated secondary sulphates. Electron probe microanalysis shows that goethite in the heaps is uraniferous, resulting from the adsorption of U(VI).

After recovery of target metals, the pregnant leach solution is neutralized to further remove metal contaminants and the resulting slurries stored in a bunded tailings pond. The activity concentrations of thorium, radium, lead and polonium isotopes are generally low in the pond owing to prior retention by secondary minerals in the heaps. However, ^{238}U activity concentrations range up to 3375 Bq/kg, which exceeds the permitted value (1000 Bq/kg) for natural radionuclides of the ^{238}U series.

1. Introduction

Considerable concerns surround the introduction of naturally occurring radioactive material (NORM) into the biosphere as a result of mining activities (Bhattacharyya, 1998). Mining and processing of uranium- and thorium-bearing ores can lead to elevated concentrations of uranium and thorium and their decay products in mine waters, tailings, waste rock and mineral dust, resulting in potential radiological and ecotoxicological risks. Natural radionuclides may leach from mining wastes to receiving water systems that are accessible to the public or used for agriculture (Darko et al., 2009). The behaviour and fate of radionuclides in the mining process needs to be understood therefore, in order to evaluate possible contamination pathways in the future. This study focuses on the behaviour of uranium and its radiotoxic daughters, ^{226}Ra, ^{210}Pb and ^{210}Po, in a bioheap leaching process applied at the Talvivaara mine in Finland.

The Talvivaara nickel deposit is located in Sotkamo, Eastern Finland. The large open-cast mine started operations in 2008. The company uses heap leaching to extract metals (Ni, Zn, Cu, Co) from a low-grade black schist ore which also contains uranium at an average concentration of 17 mg/kg (OECD, 2014). The production process at Talvivaara includes the stages of mining, crushing, heap leaching, metals recovery and removal of less valuable, residual metals (Fig. 1). The leaching solution is collected at the bottom of

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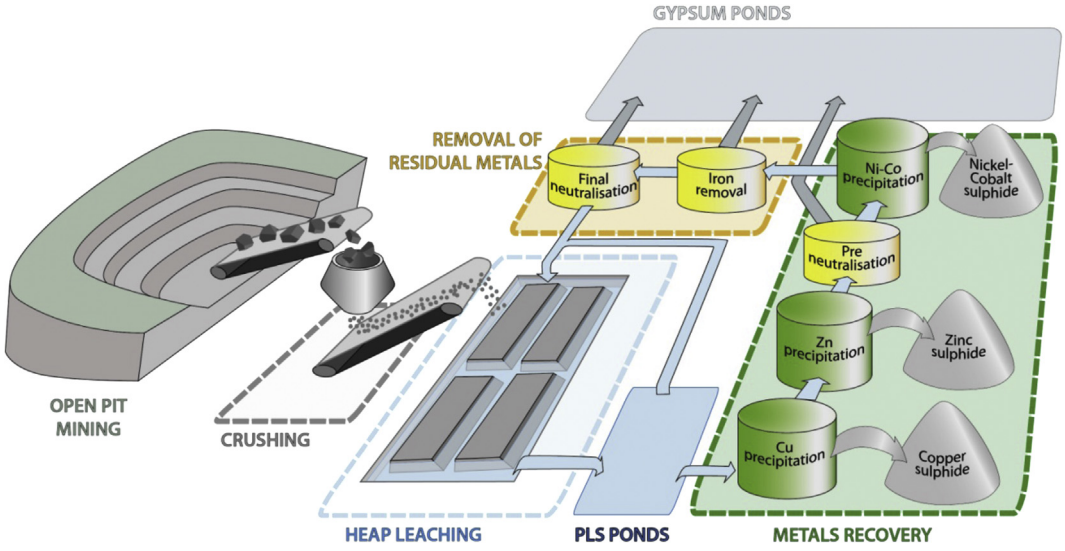

Fig. 1. Mining process at Talvivaara.

the heaps and is either re-circulated through the heap or fed to metals recovery. The most important parameters affecting the efficiency of heap leaching at Talvivaara are the particle size of the ore (<8 mm), pH of the pregnant leach solution (between 2 and 2.5) and temperature; the latter typically varying between 20 °C and 90 °C in the heaps (Pitkäjärvi, 2009). The acidity of the leaching solution is controlled by sulphuric acid and leaching of metals to solution is catalysed by endemic bacteria (e.g. *Acidithiobacillus ferrooxidans*) in the ore (Pitkäjärvi, 2009). After two years of primary leaching, the ore is excavated and re-stacked for secondary leaching to enhance metal recovery. The secondary leaching heaps constitute the final disposal sites for spent ore.

During metals recovery, copper, zinc, nickel and cobalt are precipitated from the pregnant leach solution and filtered to produce saleable metal products. After the target metals have been recovered, the solution is further purified to remove unwanted metals and returned to irrigate the heaps. The pH of the pregnant leach solution (PLS) is raised to 9–10 using lime slurry as a result of which residual metals (Mn, Mg, Fe) are precipitated as hydroxides along with gypsum. The slurry is thickened and the thickener underflow is directed to pond storage (Talvivaaran Kaivososakeyhtiö Oyj 2012, Lapin Vesitutkimus Oy, 2012). In 2010, Talvivaara announced plans to recover uranium as a by-product, given that a large proportion of the uranium originally present in the ore dissolves in the pregnant leach solution during heap leaching. Currently, uranium is mostly found in the gypsum pond tailings and, to an extent, in the Ni–Co sulphide concentrate product consigned to the Norilsk Nickel refinery at Harjavalta (OECD, 2014).

At other sites where in situ-leaching is used to recover metals, blockage of flow paths by jarosite has been a problem, especially where the pyrite content of the ore is high (Kaiman, 1978). Jarosite can be an important host for several environmental contaminants and has been shown to concentrate radium and lead (Kaiman, 1978). The same could be expected to occur in the Talvivaara heaps. Although the leaching process oxidizes U(IV) to U(VI) in the form of the uranyl ion (UO_2^{2+}) at low pH, co-precipitation of uranium decay series radionuclides with sulphate minerals is a possible retention mechanism for these nuclides in the heaps. The mobilization of radionuclides from solids to the liquid phase may occur after disposal, even a long time after rehabilitation has been completed. Therefore, determining the processes that effect radionuclide mobility at mining sites is crucial in order to first select appropriate remediation techniques and to avoid legacy environmental problems in the future.

2. Geological setting

2.1. Site description

2.1.1. Physiography

The Talvivaara open pit mine occupies an area of 60 km^2 and is located in Sotkamo, Eastern Finland (63° 58′ 30″ N, 28° 0′ 30″ E, WGS84). The surrounding landscape is typical of the region with alternating swamps, ponds, lakes and forests (Pöyry Finland Oy, 2011). The mining area straddles the watersheds of two rivers, the Vuoksi and Oulujoki, and waters in both are affected by mine drainage. The pH of small lakes and streams near Talvivaara is low and the pH buffering capacity of the local geology is limited. Consequently, dissolved metal concentrations tend to be high (Pöyry Finland Oy 2013).

2.1.2. Ore characteristics

The Talvivaara Ni-Zn-Cu-Co ore deposit is hosted in black schist of the Palaeoproterozoic Kainuu Schist Belt (Fig. 2). The deposit, discovered by the Geological Survey of Finland (GTK) in 1977, is composed of two ore bodies: Kuusilampi (2.8 km long and 40–600 m wide) and Kolmisoppi (2.5 km long and 30–350 m wide). The ore bodies are connected in the sub-surface and the maximum depth reached by drilling is 800 m (Loukola-Ruskeeniemi and Lahtinen, 2013).

Graphitic-sulphidic metasedimentary units in Talvivaara are strongly enriched in redox sensitive elements (Fe, Mo, Sb, U) and base metals (Cu, Zn, Ni) relative to average upper crustal values. Metal enrichment took place 2.0–1.9 Ga ago in a stratified oxic–anoxic, restricted marine basin, which was characterised by a high sedimentation rate of organic matter, leading to anoxic conditions when organic material was preserved in bottom sediments. The black shales of Talvivaara were deformed and metamorphosed (lower amphibolite facies) during the 1.91–1.78 Ga Svecofennian orogeny (Kontinen, 2012).