

Arsenic and antimony speciation of historical roaster waste samples from the Giant Mine, Yellowknife, Canada

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The Giant Mine has a legacy of arsenic contamination from mining and processing of gold-bearing arsenopyrite. Approximately 237,000 tonnes of As₂O₃ roaster waste (ATRW) were generated from 1952-1999 and stored in underground chambers. Knowledge of the waste geochemistry and mineralogy is essential for assessing its long-term stability and options for permanent remediation strategies. Previous studies hypothesized that Sb substitution for As in ATRW solids may affect ATRW geochemical characteristics and environmental behavior. Here, we examine ATRW samples from 9 of the 15 underground chambers to assess compositional heterogeneity and As and Sb speciation and coordination.

Bulk geochemical analyses indicate As dominates the ATRW with a wide range of Fe, Sb, S, Al, K and Pb concentrations among samples, and subtle variations in Mg, Ca, Na and Cu. X-ray diffraction and energy dispersive spectra analyses reveal that the waste is dominated by arsenolite [As₂O₃], with minor to accessory Fe oxides (maghemite, magnetite, hematite, magnesioferrite), arsenopyrite, muscovite, clinocllore, quartz, gypsum, dolomite, and other phases that likely include scorodite [FeAsO₄·2H₂O], arseniosiderite [Ca₂Fe₃O₂(AsO₄)₃·3H₂O], and yukonite [Ca₇Fe₁₂(AsO₄)₁₀(OH)₂O·15H₂O]. The majority of As and Sb is hosted by arsenolite, with some As present in other phases. Arsenic K-edge XANES spectra indicate As(III) is dominant with varying proportions of As(V) and As(-I). All samples contain variable proportions of Sb(III) and Sb(V). The As K-edge EXAFS spectra are consistent with the local structure of arsenolite plus inner sphere complexation of As with Fe oxides. Sb K-edge EXAFS spectra are dominated by Sb-O, Sb-Fe and Sb-As bonds indicative of multiple solid-phase Sb hosts including senarmontite [Sb₂O₃], tripuhyite [FeSbO₄] and (As,Sb)₂O₃.

Although these ATRW samples are dominated by arsenolite, the observed variability in solid phase mineralogy, oxidation state and molecular structure of As and Sb indicates the presence of distinct mineral phases whose geochemical behavior differs from arsenolite, an important consideration in assessing alternate chemical stabilization approaches. This study improves current knowledge of chemical heterogeneity within the chambers. It also sheds light on the As and Sb speciation and coordination environment in the ATRW, needed to predict relative solid-phase stabilities within the chambers and assess mobilization potential in long-term ATRW management.