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Mineralogical controls on antimony and arsenic mobility during tetrahedrite-tennantite weathering at historic mine sites Špania Dolina-Piesky and Ľubietová-Svätodušná, Slovakia

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Abstract

The legacy of copper (Cu) mining at Špania Dolina-Piesky and Ľubietová-Svätodušná (central Slovakia) is waste rock and soil, surface waters, and groundwaters contaminated with antimony (Sb), arsenic (As), Cu, and other metals. Copper ore is hosted in chalcopyrite ($CuFeS_2$) and sulfosalt solid-solution tetrahedrite-tennantite $\{Cu_6[Cu_4(Fe,Zn)_2]Sb_4S_{13}–Cu_6[Cu_4(Fe,Zn)_2]As_4S_{13}\}$ that show widespread oxidation characteristic by olive-green color secondary minerals. Tetrahedrite-tennantite can be a significant source of As and Sb contamination. Synchrotron-based μ-XRD, μ-XRF, and μ-XANES combined with electron microprobe analyses have been used to determine the mineralogy, chemical composition, element distribution, and Sb speciation in tetrahedrite-tennantite oxidation products in waste rock. Our results show that the mobility of Sb is limited by the formation of oxidation products such as tripuhyite and roméite group mineral containing 36.54 wt% Sb for samples where the primary mineral chemical composition is close to tetrahedrite end-member. Antimony *K*-edge μ-XANES spectra of these oxidation products indicate that the predominant Sb oxidation state is 5^+. Arsenic and Cu are also hosted by amorphous phases containing 6.23 wt% Sb on average and these are intergrown with tripuhyite and roméite. Antimony in this environment is not very mobile, meaning it is not easily released from solid phases to water, especially compared to As, Cu, and S. For samples where the primary sulfosalt is close to tennantite composition, the oxidation products associated with tennantite relicts contain 2.43 wt% Sb and are amorphous. The variable solubility of the secondary minerals that have been identified is expected to influence mobility of Sb and As in near-surface environment.

Keywords: Tetrahedrite-tennantite weathering, waste rock, antimony, arsenic, supergene minerals, tripuhyite, roméite