

Contaminant capture in anthropogenic and experimentally produced tufas - potential for legacy industrial waste treatment

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Secondary mineralisation in legacy industrial waste can produce anthropogenic tufas, i.e., surface freshwater limestones (CaCO₃) which precipitate through atmospheric CO₂ absorption into hyperalkaline (pH >10), Ca-rich leachate [1]. Metal contaminants can be captured during the mineralisation through chemisorption processes. This work studied geochemistry of anthropogenic tufas in central Scotland and subsequently recreated tufa-precipitating mechanism in the laboratory to explore its metal capture potential. The experimental results aim to promote the development of engineered tufas as a low-cost, sustainable remediation for hazardous metal contamination of surface waters at industrial waste settings.

The field sites are complex chemical microenvironments. Leachate chemistry is dominated by Ca, K, Na and S, but the diversity of source material creates strong variability in minor metal proportions between sites. Both single-point and multidirectional inflow, irregular distributive outflow over the deposit and periodic breaks in deposition during dry periods result in semi-continuous and isolated tufa deposits of distinctive chemistries, morphologies, sedimentary characteristics and colour across each site. XRD analysis revealed tufa composition of 98-100% calcite, suggesting that the metals are captured directly into calcite rather than separate metal mineral phases.

Engineered tufa precipitation was tested in batch and column experiments utilising CO₂ bubbling through hyperalkaline CaCl₂ + 2NH₄OH solution [2]. Column experiments also tested the extent of solid CaCO₃ coating of a range of surface materials. The thickest (≤0.5 mm) coating formed on materials with large surface area and roughness which slowed solution flow, thus extending reaction time for CaCO₃ precipitation in the column. Later experiments investigated Cu, Ni, Sr and Zn capture by the engineered tufa, one metal per experiment. Precipitate changed from white to blue (Cu) and yellow (Ni), most likely metal carbonates, which indicates that after reactants for CaCO₃ precipitation are exhausted, metal capture from solution may continue through other metal minerals precipitation.

[1] H. Pullin, A. W. Bray, I. T. Burke, D. D. Muir, D. J. Sapsford, W. M. Mayes and P. Renforth (2019), *Environ. Sci. Technol.* 53, 9502–9511.

[2] C. J. Grimes, T. Hardcastle, M. S. Manga, T. Mahmud and D. W. York (2020), *Crystal Growth & Design* 20, 5572–5582.