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# Toward a permanent solution to legacy As pollution: Upcycling As-rich mining waste to form metallic As(0)

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Decades of poorly-regulated mining activities have resulted in ubiquitous legacy sites polluted with intense levels of carcinogenic arsenic (As). Prominent examples include Giant Mine (Yellowknife, Canada) and the Salsigne gold mine (Aude, France), which store a combined >300,000 metric tons of As-rich waste. New lithium mining also risks producing high As waste quantities. Current methods to remediate such wastes often rely on immobilization-based approaches, including chemical stabilization, which require long-term monitoring and do not view As as a resource. However, the societal value of As is rapidly changing, with multiple regions classifying As as a critical raw material (CRM) due to the use of metallic As(0) in products needed to transition away from fossil fuels. Upcycling As waste to produce metallic As(0) used in industry thus represents a permanent solution to legacy As pollution that might also yield benefits to the same local communities affected by the waste.

Here, we demonstrate metallic As(0) forms readily from various solid mine wastes via a two-stage upcycling process consisting of 1) aqueous As dissolution and 2) reductive As(0) precipitation. We found that up to >99% of the solid-phase As in raw wastes dissolved in both pure water and 0.1 M NaOH (Figure 1), though NaOH increased As dissolution kinetics and also enhanced the dissolution of antimony (Sb) contained in the raw waste. Dosing thiourea dioxide (TDO) reductively precipitated up to >99% of the aqueous As, yielding As(0) with a purity >99% As in optimal conditions. However, aqueous Sb released from the raw waste was co-removed by TDO, which decreased the As(0) purity, indicating a trade-off in the use of NaOH to dissolve wastes with high Sb contents. Notably, synchrotron X-ray characterization (Figure 2) showed that upcycled As(0) exhibited an amorphous structure that varied dramatically from commercial crystalline As(0). The unique amorphous structure of upcycled As(0) can be leveraged for advanced manufacturing (e.g., As-bearing semiconductors), which is an active area of our research. This proof-of-concept study can initiate a transformative shift in legacy As waste management and sets the stage for future work to implement As upcycling at Giant Mine and Salsigne.

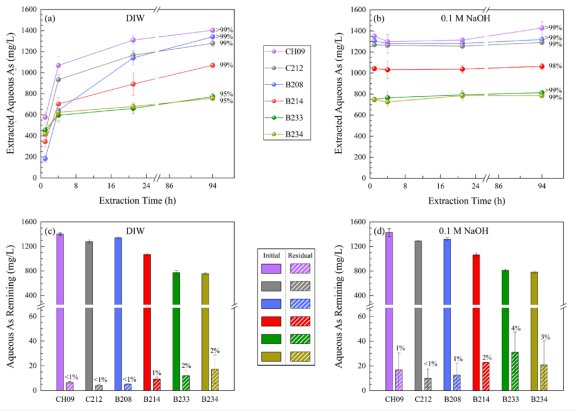



Figure 1. Aqueous As extraction from mining waste and subsequent removal of via reductive As precipitation. Data are plotted for extraction experiments using pure water (DIW) and 0.1 M NaOH at a solid-to-liquid ratio of 2 g/L over reaction times of 1-94 h. The symbols with different colors represent different raw mining waste samples from waste storage chambers of Giant Mine (CH09, C212, B208, B214, B233, B234). The total percentage of As dissolved from the raw waste is given in percentage adjacent to data points in (a, b). Data are plotted for reductive As precipitation experiments at a constant TDO/As molar ratio of 10 (c), residual As fractions are listed in % for the experiments in (c, d).

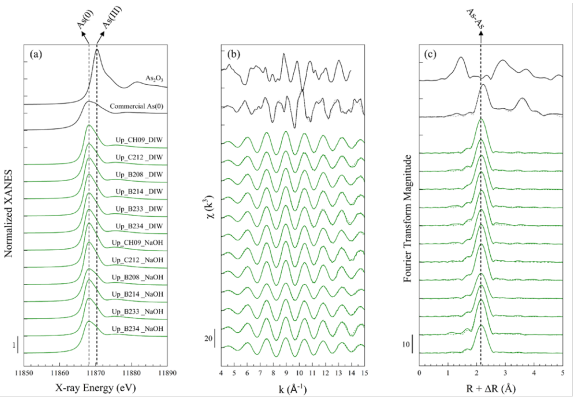



Fig. 2. Structural characterization of upcycled As(0). As K-edge XAS data of upcycled As(0) samples derived from mining waste are compared to commercial As(0) in (a-c). For sample naming, Up_C212_NaOH denotes upcycled As(0) produced via reductive As precipitation from the C212 extraction solution using 0.1 M NaOH.