An Interdisciplinary Response to Mine Water Challenges - Sui, Sun & Wang (eds)

The Removal of Arsenic(III) from Acid Mine Drainage by Mineral Trap of Tooeleite ($Fe_6(AsO_3)_4SO_4(OH)_4{\cdot}4H_2O$)

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Abstract The remediation with high concentration of As(III) adopted Mineral Synthetic Immobilization Technology to remediate acid mine drainage and provided a novel pathway of removing As(III) in form of tooeleite ($Fe_6(AsO_3)_4SO_4(OH)_4{\cdot}4H_2O$). Results showed that the low pH environment benefited the formation of tooeleite. The scanning electron microscope images showed the morphology of the mineral. The result of infrared spectroscopy revealed that the AsO_3 unit exited in the tooeleite structure and X-ray photoelectron spectroscopy confirmed that the valence of arsenic is +3. The arsenic release from tooeleite under acidithiobacillus ferrooxidans condition measured for the stability evaluation.

Keyword arsenic, acid mine drainage, tooeleite

Introduction

Acid mine drainage(AMD), which is caused by the biological oxidation of sulfidic materials, and frequently contains arsenic in the form of arsenite (AsO_3^{3-}), and/or arsenate(AsO_4^{3-}). Tooeleite ($Fe_6(AsO_3)_4SO_4(OH)_4{\cdot}H_2O$) is only ferric arsenite sulphate in acid mine drainage (AMD) (Cesbron 1992), tooeleite was proposed as arsenic mineral storage for remediation of metallurgical waste water (Liu 2013). Tooeleite is the common As (III)-loaded secondary mineral in AMD, which controls the arsenic release and migration. Mineral Synthetic Immobilization Technology (MSIT) is an innovative technology, which can solidify contaminants as mineral pattern.

Acidithiobacillus ferrooxidans is common autotrophic bacteria in AMD, which can catalyze the oxidation of Fe(II) into Fe(III) and enhance the formation of some typical secondary arsenic containing minerals in AMD (Duquesne 2003). The aim of this paper is to report the dissolution characteristic of tooeleite with acidithiobacillus ferrooxidans related to arsenic release.

Materials and methods

Tooeleite synthesis. The mineral tooeleite was synthesized based on our previous reported. Mixed 1 L acidic 0.1335 mol/L As (III) solution (adjust pH=1.3 by 1 mol/L H_2SO_4) with 1 L equivalent molar concentration Fe (III) solution in constant-temperature double-layer glass reaction kettle at 95℃, the pH of mixed solution was adjusted to pH=2 by 1 mol/L NaOH using peristaltic pump at 5 mL/min flow rate. The suspension solution was aged and agitated at 50 rpm under 95℃ for 2 h, and then filtered by 0.25 μm membrane. The precipitate was washed by distilled water and dried at 80℃ for one day.

Bacterial strain. Acidithiobacillus ferrooxidans used in this study was developed in 9K medium (KCl 0.1 g/L, $(NH_4)_2SO_4$ 3.0 g/L, K_2HPO_4 0.5 g/L, $MgSO_4{\cdot}7H_2O$, 0.5 g/L, $Ca(NO_3)_2$, 0.01 g/L), in which ferrous ion is the energy source. The pH of medium was adjusted to pH 2.0 ~ 3.0. Tooeleite sample of 2.5 g was added to 200 mL bacterial culture after it was incubated at 30℃ and 240 rpm for 10 days. For biotic and abiotic comparison, the equivalent tooeleite sample was also added to only 9K medium. These suspension solutions were

continuously oscillated at 240 rpm and 30 ℃ for 30 days. The final dissolution products were dried in 80 ℃ for 24 hours.

Result and discussion

SEM of tooeleite

Tungsten filament scanning electron microscopy (SEM) used EVO 18 of ZEISS which made in German. Fig.1 shows the morphology of the mineral tooeleite. Mineral crystal plate shaped and the size of each cluster is about 1 μm.

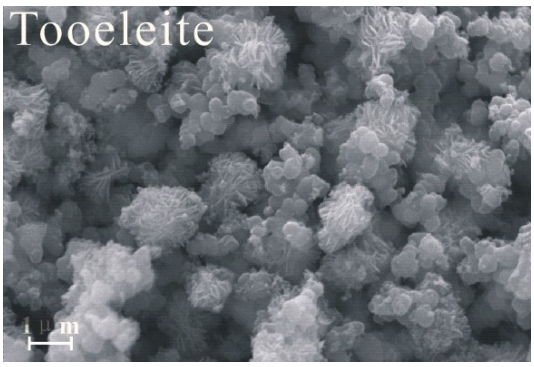

Fig. 1 *SEM of tooeleite*

XPS of tooeleite

X-ray photoelectron spectroscopy was texted by ESCALAB 250 of ThermoFisher Scientific. The infrared spectrum of arsenite of tooeleite occurs in the 400-900 cm^{-1} spectral range, as illustrated in fig. 2. In fig. 2, an obvious peak is observed with binding energy of 44.05 (eV), thus indicating that the As valence in tooeleite is +3.

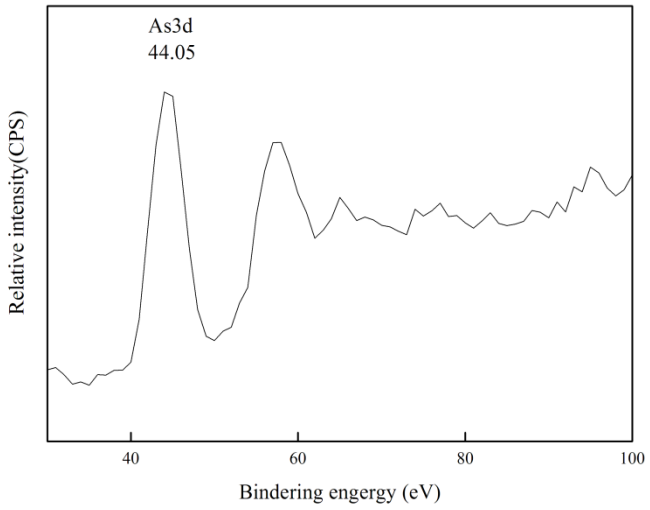

Fig. 2 *XPS of tooeleite*

Infrared spectroscopy

Infrared spectra of mineral were collected using a Nicolet Nexus870 FTIR spectrometer. Spectra over the 400-4000 cm^{-1} range were obtained by the co-addition of 64 scans with a resolution of 4 cm^{-1} and a mirror velocity of 0.6329 cm/s. The degenerate mode of SO_4^{2-} of tooeleite crystal is found to be split into the band at 1109cm^{-1} (fig.3), although the vibrational mode theoretically occurs at 1105 cm^{-1}.

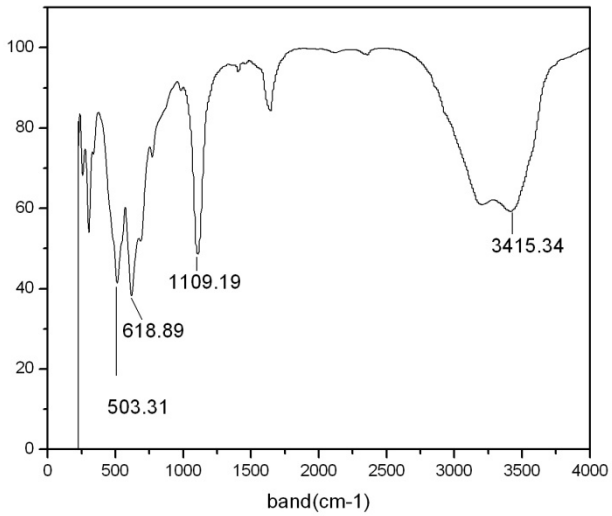

Fig. 3 *Infrared spectroscopy of tooeleite*

Arsenic release of tooeleite during 30 days is illustrated in fig. 4. The arsenic release of tooeleite in medium has decreased tendency at early 2 days, which probably due to refixation of arsenic on surface. The arsenic release gradually increases after 2 days and up to 100 mg/L, which is at lease 104 times than drinking quality for arsenic requirement (10 μg/L) (Martinez 2011). Once tooeleite contacts with acidithiobacillus ferrooxidans, arsenic release is promoted obviously and as high as 345 mg/L finally, which is 3 times than abiotic environment. Meanwhile, arsenic release is higher in the bacterial-favorable environment (pH=2) than critical environment (pH=4), indicating the bacterial activity also is a practical consideration for field study.

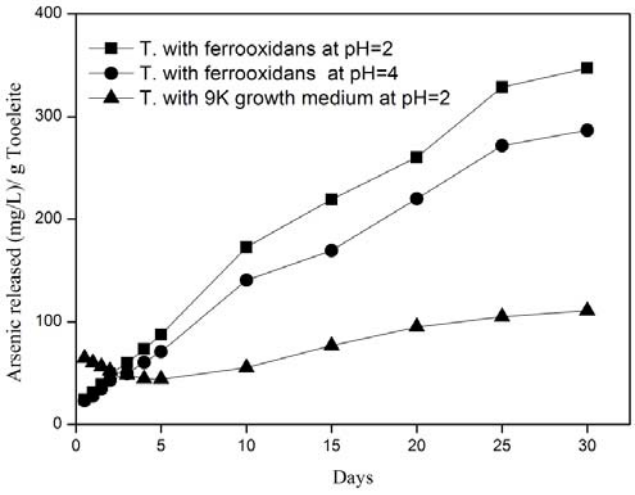

Fig. 4 *Arsenic released from tooeleite with time under ferrooxidans culture and medium at pH=2 and 4*

Conclusions

The release of arsenic from tooeleite and its stability with acidithiobacillus ferrooxidans were evaluated based on the batch experiments for 30 days. Once tooeleite contacts with acidithiobacillus ferrooxidans, arsenic release is promoted obviously, which is 3 times than abiotic environment. Thiobacillus ferrooxidans can promote the dissolution of tooeleite and arsenic release as compared to alone tooeleite.

Acknowledgments

This work was supported by National Natural Science Foundation of China (No. 41372164), and the Sixth "China-South Africa" Joint Research Plan of Ministry of Science and Technology of China (2012DFG71060).

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