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Thermally-induced structural reorganisation of iron-arsenic minerals during bushfires

MARJANA YEASMIN[1], DANE LAMB[1], SEPIDE ABBASI[2], ANDREW R. C. GRIGG[3], USMAN FARYAD[1], EDWARD D. BURTON[4], ANGELIA SEYFFERTH[5], SUZIE M. REICHMAN[6] AND RUBEN KRETZSCHMAR[3]

[1]RMIT University
[2]ANSTO
[3]ETH Zurich
[4]Southern Cross University
[5]Arizona State University
[6]The University of Melbourne

Climate change is expected to increase bushfire frequency, raising concerns about arsenic (As) cycling and changes in global exposure patterns. Elevated soil temperatures during bushfires can alter the chemical composition and stability of minerals, potentially producing and releasing toxic forms of As. Soil organic matter plays a critical role in changing the redox dynamics of As during bushfires through provision of functional groups and promoting electron shuttling during heating. This study investigates the geochemical response of As^{III} and As^{V}-bearing iron (Fe) minerals under simulated bushfire conditions. In this regard, we focus on the structural reorganisation of tooeleite ($Fe^{III}_6(As^{III}O_3)_4SO_4(OH)_4 \cdot 4H_2O$) and scorodite ($Fe^{III}As^{V}O_4 \cdot {}_2H_2O$) during bushfire-relevant heating regimes (ranging from 100-350°C) in the absence and presence of organic substrates (in the form of citric acid and humic acid at a 5% C (w/w) ratio).

X-ray diffraction revealed a heating-induced loss of crystallinity starting at 175°C for tooeleite and 275°C for scorodite. Thermally-altered scorodite exhibited a Mössbauer spectroscopic blocking temperature of 5-77 K, while thermally-altered tooeleite remained unordered even at 5 K. Heating tooeleite at 350°C caused XRD amorphisation, with Mossbauer spectroscopy showing 6-9% reduction of Fe^{III} to Fe^{II} and broad doublets (at 77 K) that were inconsistent with known nano-crystalline Fe phases. For scorodite at 350°C, Mössbauer spectroscopy (at 77 K) identified a mixed mineral phase with a broad Fe^{III} doublet, consistent with ferrihydrite, and another Fe^{III} doublet, which could be an amorphous ferric arsenate phase, indicating significant mineralogical alterations. Additionally, arsenic K-edge XANES and XPS revealed extensive As^{III} oxidation of tooeleite at 350°C, which was inhibited by humic acid and produced 8% As^{II} upon heating, while scorodite displayed up to 13% reduction of As^{V} to As^{III} in the presence of organic acids. Iron K-edge and As K-edge EXAFS of tooeleite confirmed the formation of a collapsed mineral structure, while scorodite shows the formation of ferrihydrite and AFA. These findings suggest that bushfire heating triggers complex Fe-As transformations, breaking down crystalline phases and creating new nano-crystalline forms.