Supporting Information for:

Field-based accounting of CO_2 sequestration in ultramafic mine wastes using portable X-ray diffraction

Connor C. Turvey[1]*, Siobhan A. Wilson[1], Jessica L. Hamilton[1], Gordon Southam[2]

[1]School of Earth, Atmosphere & Environment, Monash University, Clayton, Melbourne, Victoria 3800, Australia

[2]School of Earth Sciences, The University of Queensland, St Lucia, Queensland 4072, Australia

* Corresponding author, connor.turvey@monash.edu, +61 03 9905 4382

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1. Description of Mineral Standards

The chrysotile standard used to prepare the artificial tailings samples was a picrolite from the Clinton Creek Chrysotile Mine, Yukon, Canada [previously studied by Wilson et al.(2006)]. Hydromagnesite was obtained from a carbonate playa near Atlin, British Columbia, Canada (described by Power et al.(2014)). The pyroaurite standard was made by conversion of iowaite [$Mg_6Fe_2(Cl)_2(OH)_{16}\cdot 4H_2O$] from the Mount Keith Nickel Mine, Western Australia. The iowaite was placed in an excess of deionised water in a vigorously stirred beaker for 48 hours. This produces pyroaurite through anion exchange (Bish, 1980; Miyata, 1983) using atmospheric CO_2 gas to generate the dissolved inorganic carbon required for reaction. X-ray Diffraction (XRD) analysis of the pyroaurite standard showed that it contained pyroaurite, brucite and minor amounts of unreacted iowaite. Brucite was present as an impurity in the iowaite from the Mount Keith Nickel Mine. The relative abundances of pyroaurite and brucite in the standard were determined to be 60.1 and 39.3 wt.%, respectively, by Rietveld refinement of XRD data. The magnetite standard was obtained from a commercial supplier and was found to be 94.4% pure, with minor hematite contamination (5.6 wt.%).

2a. Instrument Details: inXitu Terra

XRD patterns were collected using an inXitu Terra portable XRD based in the School of Earth, Atmosphere and Environment at Monash University. The instrument was equipped with a Co X-ray tube. Patterns were collected over a 2θ range of 5−55°. The inXitu Terra is designed to acquire data over the entire angular range with each exposure. Multiple analysis times were trialled to assess the capabilities of the instrument. For the synthetic and natural tailings samples a total of 590 exposures were used for a total analysis time of 128 minutes per sample. Mineral identification from all patterns was performed using DIFFRAC.EVA V.2 (Bruker

AXS) with reference to standard patterns from the International Center for Diffraction Data Powder Diffraction File (PDF-2) database and the Crystallography Open Database (COD).

2b. Instrument Details: Bruker D8 Advance

XRD patterns were collected using a Bruker D8 Advance X-ray Diffractometer in the Monash X-ray Analytical Platform. The instrument was equipped with a LynxEye 1D Position Sensitive Detector Data acquisition was done using a Cu X-ray tube, operated at 40 kV and 40 mA, over a 2θ range of 3–80° with a step size of 0.02°/step and a dwell time of 1 s/step, resulting in analysis time of 65 minutes. Mineral identification from all patterns was performed using DIFFRAC.EVA V.2 (Bruker AXS) with reference to standard patterns from the International Center for Diffraction Data Powder Diffraction File (PDF-2) database and the Crystallography Open Database (COD).

3a. Refinement Strategy: inXitu Terra

The most accurate refinement strategy for XRD patterns collected using the inXitu Terra consisted of three stages. Firstly, the scale factors and unit cell parameters of major and minor phases present in each sample were refined. Secondly, Lorentzian crystallite size and strain for magnetite, pyroaurite, iowaite and hydromagnesite were allowed to refine (from default starting values of 1000 nm and 0.1 respectively). As a last step trace phases were included in the refinement. No preferred orientation corrections were used because they did not appreciably improve the fit and refinement statistics. This is likely because the inXitu Terra agitates samples within its stage during analysis, which mitigates the adverse effects of preferred orientation. A PONKCS model of chrysotile was used to model serpentine minerals within the sample. Refinements were trialled using a PONKCS model for lizardite and models for both chrysotile and lizardite however they did not appreciably improve the accuracy of the results and so were ultimately discarded in favour of refinements that only used the chrysotile PONCKS model.

3b. Refinement Strategy: Bruker D8 Advance

XRD patterns obtained from the D8 Advance XRD were refined using four steps. First, the scale factors and unit cell parameters were refined. Secondly, the Lorentzian crystallite size and strain values for pyroaurite, iowaite, brucite and hydromagnesite were allowed to refine from default starting values of 1000 nm and 0.1 respectively. The March-Dollase preferred orientation correction (Dollase, 1986; March, 1932) was applied to pyroaurite, iowaite, brucite and hydromagnesite. Lastly, trace phases were added and their abundances were allowed to refine. A PONKCS model of chrysotile was used to model serpentine minerals within the sample. Refinements were trialled using a PONKCS model for lizardite and models for both chrysotile and lizardite however they did not appreciably improve the accuracy of the results and so were ultimately discarded in favour of refinements that only used the chrysotile PONCKS model.

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