

Hydrotalcites as a carbon sink in serpentinites

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Ultramafic mine tailings are being investigated for their potential to trap and store atmospheric CO₂ via passive carbonation reactions. Hydrotalcite minerals may be a potential carbon sink in many tailings piles. Hydrotalcites can trap atmospheric CO₂ in multiple ways, either through the carbonation of gangue brucite present in tailings or via anionic exchange reactions with non-carbonate bearing hydrotalcite minerals.

Pyroaurite [Mg₆Fe³⁺₂(CO₃)(OH)₁₆·4H₂O], a carbonate bearing hydrotalcite mineral, is found throughout the tailings pile at the Woodsreef Chrysotile Mine (New South Wales, Australia). This may represent a second carbon sink at the site, in addition to the hydrated Mg-carbonate crusts that are forming on the surface of the tailings pile.¹ It is necessary to determine whether the pyroaurite is forming by reaction of gangue minerals with atmospheric CO₂ or was already present in the ore. If the pyroaurite is sequestering atmospheric CO₂, it becomes important to quantify the abundance of this mineral in the tailings for the purpose of carbon accounting. Quantitative mineralogical results can be then be used to estimate the total amount of CO₂ sequestered at the site and to determine the relative contributions made by the different carbonate phases.

Our stable carbon and oxygen isotope results show that there are two distinct reservoirs for atmospheric CO₂ in the tailings at Woodsreef: isotopically heavier hydromagnesite and isotopically lighter pyroaurite. Stable C and O isotopes can be used to determine the source of carbonate within the structures of hydromagnesite and pyroaurite. Using mineral precipitation and anion exchange experiments, we have shown that incorporation of aqueous carbonate into the pyroaurite structure produces a small stable carbon isotope fractionation and that isotopically light δ¹³C values of pyroaurite samples instead reflect kinetic diffusion of atmospheric CO₂ into solution.

Samples were taken from across the tailings pile to determine the horizontal and vertical extent of pyroaurite formation at Woodsreef with the goal of refining our estimate of the CO₂ sequestration rate. Quantitative X-ray diffraction with the Rietveld method was used to determine the composition of tailings. The surface of the tailings, down to a depth of 2 cm is cemented with hydromagnesite [Mg₅(CO₃)₄(OH)₂·4H₂O]; however, pyroaurite is present throughout the tailings, to depths of >1 m, with higher abundances found at the surface. A poorly crystalline phase that resembles a thermal decomposition product of nesquehonite² is found deeper into the tailings (50 – 100 cm depth). There is an inverse relationship between the abundances of brucite and pyroaurite with depth (with brucite less abundant at the surface and pyroaurite more so), which is consistent with the interpretation that the pyroaurite at the site is forming by carbonation of brucite in the shallow tailings that are exposed to the atmosphere.

Modal mineralogy results, combined with tailings density data, can be used to estimate the mass of CO₂ sequestered by Mg-carbonate minerals and pyroaurite in the tailings. For a given cubic metre of tailings that are exposed to the atmosphere, 10.4 kg of CO₂ are sequestered within alteration minerals. By extrapolating from 1 m³ to the entire surface of the tailings pile we estimate that the Woodsreef Mine has sequestered ~13.8 kt of CO₂, with approximately 20% of the CO₂ sequestered within pyroaurite.

1. Oskierski, H. C.; Dlugogorski, B. Z.; Jacobsen, G., Sequestration of atmospheric CO₂ in chrysotile mine tailings of the Woodsreef Asbestos Mine, Australia: Quantitative Mineralogy, isotopic fingerprinting and carbonation rates. *Chemical Geology* **2013**.
2. Morgan, B.; Wilson, S. A.; Madsen, I. C.; Gozukara, Y. M.; Habsuda, J., Increased thermal stability of nesquehonite (MgCO₃·3H₂O) in the presence of humidity and CO₂: Implications for low-temperature CO₂ storage. *International Journal of Greenhouse Gas Control* **2015**, *39*, 366-376.