

Characterisation of ultramafic tailings and their alteration: From process control to carbonation potential

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Ultramafic tailings represent viable feedstocks for CO₂ mineralisation, providing a storage capacity of about 250 Mt CO₂/year in 2024^[1]. To facilitate realisation of this capacity, tailings characterisation focused on process control can be modified to provide valuable insights into alteration and associated changes in carbonation potential of ultramafic tailings.

Geochemical process control data commonly include divalent cation concentrations for calculating the maximum theoretical carbonation potential, with R_{CO_2} of the studied tailings between 2.10 and 2.44 ($n = 97$). The olivine-pyroxene elemental ratio (OPE)^[2], calculated from major element data, varies from 1.35 to 1.64, consistent with an olivine-bearing protolith. Major element ratios constrain the degree of weathering, which is associated with preferential brucite dissolution, removal of MgO and lowering of carbonation potential. MgO/SiO₂ and Al₂O₃/SiO₂ in agreement with the terrestrial mantle array confirm that the investigated tailings are not affected by weathering.

However, major element compositions do not capture the otherwise isochemical addition of H₂O and CO₂ during serpentinisation and carbonation of the ultramafic protolith, respectively. This requires the determination of volatile contents, indicating that the tailings have undergone partial serpentinisation (average 5.5 wt.% H₂O) and carbonation (average 3.0 wt.% CO₂). Serpentinisation of olivine-rich protoliths with high OPE may increase carbonation potential through formation of reactive brucite. Carbonation, on the other hand, consumes brucite and naturally adds CO₂ to tailings that may be lost during subsequent industrial carbonation.

Quantitative mineralogy records alteration of the studied tailings as decrease in olivine (47 to 10 wt.%) and increase in serpentine content (4 to 37 wt.%) during serpentinisation, or the formation of talc (8 to 22 wt.%) and/or magnesite (2 to 13 wt.%) during carbonation and enables more realistic, dissolution kinetics-informed estimates of carbon dioxide removal (CDR) potential^[1]. However, measurement uncertainties and detection limits of mineral quantification are generally higher than that of chemical composition measurements. As such, a combination of chemical and mineralogical characterisation, including volatile contents, is necessary to correctly assess alteration of ultramafic tailings and its effects on carbonation potential.

[1] Bullock et al. (2022) *Sci. Total Env.* 808, 152111.

[2] Cutts et al. (2021) *Geochem. Geophys. Geosystems* 22, e2021GC009989.