

Cellular Dissolution at Hypha- and Spore-Mineral Interface during Fungal Weathering

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Microbial weathering develops through intentional or unintended reactions between microbes/metabolites and minerals. Whereas the latter can be modeled by bulk dissolution, the former often involves complicated cell-mineral interfacial processes and hence is less understood. For fungus-mineral interaction, an additional but unique influence, i.e. the biomechanical forces, needs to be evaluated as surface-bound cells can apply physical pressure through hyphae to disrupt crystal structures. As high as 10-20 MPa turgor pressure was reported during hyphal growth (approximately 100 times that of a typical car tire), strong enough for fungi to penetrate grain boundaries and break crystalline particles along the cleavage directions. What is more unique to fungi but yet largely unknown is the relative scales of cellular dissolution associated with different cell segments in light of the turgor pressure difference between hyphae and spores. Here we examine lizardite ($\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$) dissolution by single cells of a native fungal strain (*Talaromyces flavus*) from a serpentine mine using confocal laser scanning microscopy (CLSM), atomic force microscopy (AFM), and focused ion beam transmission electron microscopy-energy dispersive X-ray spectroscopy (FIB-TEM-EDX) to explore the mechanism, driving force, and magnitude of the interfacial reactions. Bulk experiments reveal that the fungi significantly enhanced dissolution. Moreover, dissolution was substantially stronger when cell-mineral contact was permitted in comparison to the cases where the cells were separated from the minerals grains via a semi-permeable membrane, suggesting the bioweathering results from combined active and passive microbial dissolution. In addition, the fungal effect appeared to steer the dissolution to a non-stoichiometric pathway. The molar ratios of Mg to Si during abiotic dissolutions varied between ~ 2 and ~ 1.3 but mostly stayed near the theoretical value of 1.5, signaling a congruent dissolution. In contrast, the ratio during bioweathering deviated progressively more strongly from the stoichiometric value as the dissolution continued and reached ~ 4 to ~ 7 at the end of experiments, indicating either a preferential release of Mg or a re-precipitation of silica. Analyses of the cell-mineral interface show (i) significant pH reduction (~ 1 pH unit) in the vicinity of surface-bound cells upon mineral attachment, (ii) extensive occurrence of deep (~ 200 to ~ 2000 nm) channels and shallow (~ 50 nm) circular pits (features well resembling the size and shape of the hyphae and spores), (iii) exclusive Fe loss (by as much as 70%) from the mineral at the cell-mineral interfaces (i.e. in comparison to solution-mineral interfaces), and (iii) destruction of the mineral crystal structure below surface-colonized hyphae but not spores. Compared to the results from bulk experiments and at the mineral-water interface, these observations indicate (1) only attached cells release siderophores, and (2) biomechanical forces of hyphal growth are indispensable for fungal weathering and strong enough to breach the mineral lattice. Estimated mineral volume loss at the interface suggests that cellular dissolution can ultimately account for ~ 40 - 50% of the overall bioweathering, significantly larger than the previous estimate of $\sim 1\%$ contribution.

Keywords: fungal weathering, microbe-mineral interactions, lizardite, cell surface pH, interfacial reactions, siderophores