Experimental investigation of Mg-silicate-carbonate interactions during authigenesis and early diagenesis

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A number of the key carbonate successions used to interpret biological evolution, seawater geochemistry, and rift-sag basin evolution contain Al-poor sedimentary Mg-clays, such as stevensite, sepiolite, kerolite, and/or talc. The chemistry and crystal structure of these clays and their authigenic associations with carbonate minerals likely record important information about the physical and chemical conditions associated with the formation of these successions. However, at present, only broad constraints may be placed upon their formation, and relatively little is known about their interactions with the carbonate minerals with which they are associated. Furthermore, the potential effects of diagenesis on their crystallinity and chemistry have only been constrained by a handful of studies.

To fill this knowledge gap and provide the requisite data for future modeling studies, we have conducted laboratory experiments to explore how interrelated geochemical factors such as solution composition, pCO_2, pH, degree of supersaturation, temperature, and mineral substrate affect the heterogeneous precipitation of carbonates onto Mg-silicates, Mg-silicates onto carbonates, and the homogeneous nucleation and growth of both mineral groups from supersaturated solutions. We have also performed a number of parallel experiments to examine these same processes on the Mg-free, aluminous clays kaolinite and gibbsite. Experiments were performed either at fixed pH or in free-drift mode in order to enable separation of the contributions of individual processes to the overall mineral growth regime. The experiments were specifically designed to permit high resolution characterization of the solid precipitates, using high resolution x-ray diffraction, Fourier transform infra-red spectroscopy, and scanning electron microscopy. Nonetheless, changes in fluid chemistry were additionally monitored throughout. In general, the results show a sensitivity to solution Mg/Ca ratio, which appears to be more significant in substrate-free, homogeneous nucleation experiments than in the experiments containing mineral substrates. Interesting, although not yet fully constrained, forcings appear to control the relative abundance of aragonite and Mg-rich calcite precipitated during the experiments.