MOLECULAR SCALE MECHANISMS OF SOLID SOLUTION CRYSTALLISATION AND MINERAL REPLACEMENT REACTIONS.

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The formation of solid solutions, and the development of replacement reactions between minerals are important phenomena in many geological environments. Both the chemical composition of the growing solid solutions and the mechanisms of mineral replacement are strongly controlled by the processes occurring at the crystal-solution interface. Two factors are most important: (i) the supersaturation state of the solution with respect to the crystallising phases and (ii) the dissolution and growth mechanisms operating at a molecular scale on the different faces of the crystals. We present a new approach to the study of both crystallisation of solid solutions from aqueous solutions and replacement reactions based on a rigorous evaluation of the supersaturation state of the system under investigation, combined with in situ Atomic Force Microscopy (AFM) observations of the dissolution and growth mechanisms. For solid solutions both the supersaturation state of the solution and the transitional supersaturations for different growth mechanisms depend on the solid composition. Mineral replacement reactions are characterized by the evolution of the supersaturation with respect to the initial and new phase. In situ AFM observations made during the crystallisation of the solid solutions (Ba,Sr)SO4 and (Ca,Cd)CO3 from aqueous solutions, and during the transformation of phosgenite(Pb2Cl2CO3) into cerussite (PbCO3) provided information which support the crystallisation models proposed.