Effect of pH and temperature on zeolite precipitation rates and mechanisms from amorphous precursors

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The predicted precipitation of zeolites due to the dissolution of bentonite during alkaline alteration in barrier systems for the gelogical disposal of radioactive wastes may alter the properties of the barrier system. It is therefore important to elucidate the formation rates and mechanisms of zeolites over a wide range of conditions in order to predict their crystallization behavior in barrier conditions. Zeolite formation is typically preceded by an amorphous precursor, the transformation of which is seen as a rate-controlling step. However, the mechanism and rates of transformation remain poorly understood. This study focuses on the effect of pH and temperature on the transformation of the amorphous precursor into crystalline zeolite.

Batch zeolite synthesis experiments were carried out at pH ranging from 9.5 to 13.5 and at temperatures ranging from 25°C to 90°C by mixing NaOH, $NaAlO_2$ and $Na_2SiO_3.9H_2O$ at varying proportions to achieve a fluid with the composition of 0.5 M Na, 0.24 Si and 0.03 M Al. The precipitates were extracted at different times by centrifugation and freeze drying. The supernatants were analyzed using ICP-AES for Al and Na and UV-Vis for Si. The solids were examined using XRD, FTIR, Raman spectroscopy and SEM-EDX.

Results show the rapid formation of an amorphous precursor phase, followed by slower transformation into crystalline zeolite. Crystal size distribution data suggest that the dominant transformation mechanism is dissolution and reprecipitation. Higher pH and temperature promotes rapid transformation of the amorphous precursor to zeolite. Spectroscopic data indicate lower degrees of Al substitution and lower amounts of T-OH bonds in solids formed at lower pH. These results indicate that precursor phases formed at lower pH are relatively more stable and less susceptible to dissolution. Therefore, zeolite precipitation is promoted at higher pH due to the rapid dissolution of the amorphous precursor. The results of this study show that pH controls the structure and chemistry, and therefore, the stability of the zeolite precursor phases.

Archean geodynamic: Fingerprinting sagduction vs subduction processes

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The geodynamic processes responsible for the formation of Archean crust are matter of debate. Contrary to the general belief, sagduction and subduction are not incompatible processes, as both involve the deep burial and exhumation of surface and near-surface rock units. In order to better understand how to distinguish these processes, we investigated mid- to high-pressure metamorphic rocks from two key Early Archean localities: i) the East Pilbara Granite-Greenstone Terrane (EPGGT, Western Australia), considered as a reference model for sagduction [1], and ii) the Barberton Greenstone Belt (BGB, South Africa) either described in terms of subduction [2] or sagduction [3]. These two terranes display narrow belt of greenstone (ultramafic and mafic metabasalts and minor metasedimentary rocks) in association with broad TTG (tonalite-trondhjemite-granodiorite) granitoids. Estimates of the rate of heating/cooling and burial/exhumation of these granite-greenstone belts were evaluated using a combination of structural, metamorphic, and geochronological data coupled with numerical simulations.

In the EPGGT, we confirm that burial and exhumation of cold and dense greenstones is linked in time with crustal melting and granitoid dome formation. Thermobarometric, geochronological and numerical constraints indicate that metamorphism and deformation occurred within less than 10 Ma, thus implying burial and exhumation rates of some centimeters per year. The same approach applied to the BGB yielded a more complex and probably polymetamorphic history and suggest a possible different scenario. Burial/exhumation rates are possibly one order of magnitude slower and more progressive with respect to EPGGT, implying longer process.

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