

Experimental hydrothermal alteration of partially metamict zircon

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ABSTRACT

We present the results of a series of hydrothermal experiments on grains from two partially metamict zircon samples from Sri Lanka in the temperature range 350 to 650 °C and with different solutions (2 M AlCl₃, 2 M CaCl₂, pure H₂O, and a multi-cation solution). Under these conditions, sharply bounded reaction fronts penetrated into the zircon grains and developed complex lobate and rim structures that resemble structures found in natural zircon systems. The reaction zones are characterized by a marked increase in the cathodoluminescence intensity, a decrease of the back-scattered electron emission, and an increased degree of structural order, as revealed by micro-Raman and infrared spectroscopy. Sensitive high-resolution ion microprobe and electron microprobe measurements revealed that the altered areas gained solvent cations (e.g., Ca²⁺, Ba²⁺, Mg²⁺, Al³⁺) from the solution and lost variable amounts of Zr, Si, Hf, the REE, U, Th as well as radiogenic Pb. A comparison between “dry” and “hydrothermal” annealing trends shows that the kinetics of structural recovery, including recrystallization of the amorphous phase in metamict zircon, is strongly enhanced under hydrothermal conditions. This finding suggests that water “catalyzes” structural recovery processes in metamict zircon. We found that the structure of the reacted areas does not resemble that of crystalline zircon, i.e., is still characterized by a temperature-dependent degree of disorder, which would not be expected if the reaction is controlled by a coupled dissolution and re-precipitation process. Instead, the alteration process can be described best by a diffusion-reaction-recrystallization model. In this model, it is postulated that the diffusion of water into the metamict structure is the driving force for moving recrystallization fronts. We found that the rate and the extent of solid-state recrystallization of the amorphous phase is an important factor in determining the mobility of trace elements. This interpretation is indicated by the observation that trace elements, including U and Th, were preferentially lost during the reaction with a fluid at low temperatures, where recrystallization of the amorphous material was slow or not activated at all. The observed chemical alteration patterns are believed to reflect a competition between the kinetics of long-range diffusion and ion exchange and the kinetics of the short-range diffusion necessary for the recrystallization process.