PATTERN FORMATION IN METAMICT ZIRCON UNDER HYDROTHERMAL CONDITIONS

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Spatial self-organization is extremely widespread in nature. Most important is the formation of precipitation patterns in the wake of moving reaction fronts (known as Liesegang phenomenon), resulting, e.g., from the diffusion of a species A in a gel with a species B where the reaction between A and B forms a precipitate C. The instabilities underlying this phenomenon are the result of a competition between the kinetics of diffusion and reaction processes. Treatment of an almost fully amorphous zircon in hydrothermal solutions (e.g., 195 h/0.1N HCl/1 kbar/400°C) produced interesting irregularly curved or oscillatory curved patterns of spatial self-organization, as seen in cathodoluminescence (CL) and backscattered electron (BSE) images. A regular spacing between different layers does not agree with the spacing law seen in Liesegang phenomena, suggesting a different mechanism. The chemical and structural nature of these non-equilibrium patterns has been investigated by electron and ion microprobe and micro-Raman and IR spectroscopy to first derive a qualitative understanding of the pattern forming processes, which should be tested by computer modeling in future work. Micro-Raman measurements indicate that the bright-CL/BSE layers consist of almost pure well-ordered zircon, whereas monoclinic zirconia could additionally be detected inside the dark-CL/BSE regions. These areas also contain lower Zr concentrations and higher amounts of water than the bright-CL/BSE regions, as shown by electron and ion microprobe mapping, respectively. Powder IR spectroscopic measurements of completely hydrothermally reacted zircon powders from the same zircon indicate the occurrence of significant amounts of residual amorphous material. It is proposed that the diffusion of water into the amorphous network catalyzed recrystallization and was the driving force for moving reaction fronts. Since crystalline zircon
can structurally dissolve only a limited amount of hydroxyl, any excess water had to be expelled from the newly grown crystalline sites into the remaining amorphous areas. It is hypothesized that the increasing water content in the amorphous areas eventually has locally stabilized the amorphous state and caused partial decomposition of amorphous zircon to zirconia and silica. The competition between the catalytic and stabilizing effect of water may thus have produced instabilities that caused the observed self-organization patterns.