

## Formation of Liesegang rings in borosilicate glass during experimental alteration

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Borosilicate glasses are currently the standard material for the immobilization of high-level nuclear waste. Numerous studies have been carried out to study the aqueous durability of such glasses, which have indicated that under ambient conditions the corrosion of the glass is initially controlled by ion exchange and later by hydrolysis of the glass network, forming a so-called "leached layer", before the glass dissolves. For the present study we have synthesized a borosilicate glass with a complex composition, including network modifiers such as Na, Ce, Al, Mn, Li, Ca, Mg, and Ti (WAK glass), from which we cut cuboids with edge lengths of about 2.6 mm. These cuboids were treated in a 1 M HCl solution at temperatures between 100 and 220 °C for 4 days. For one experiment (at 150°C) the solution was enriched with 47.5 at.% <sup>18</sup>O. After the experiments, the formerly translucent, yellowish glass cuboids showed a milky-white color, but their external shape has not changed, as in pseudomorphism. Interestingly, the measured weight and elemental loss values increase regularly from 100 to 150°C, and then decrease towards a local minimum between 170 to 190°C before they increase again. Backscattered electron images on cut glass samples revealed reaction rims ("leached layers") with complex oscillatory banding structures and bifurcations (Liesegang rings), which form a layer between the unaltered glass and an unpatterned outer rim. Whereas the outer rim consists of almost pure SiO<sub>2</sub>, the patterned layer retained some Ce, Ti, and Al, located now within distinct bands. The occurrence of Liesegang-like banding structures suggests, at a first glance, a diffusion-controlled alteration process. However, infrared spectroscopic measurements on the alteration rim formed in the <sup>18</sup>O-enriched solution show a large mass shift of the Si-O vibrational modes (even close to the interface between the unaltered glass and the alteration layer) when compared with measurements on the alteration layer formed in "normal" <sup>16</sup>O-enriched solution. This indicates a high concentration of <sup>18</sup>O throughout the alteration rim. Furthermore, the chemical interface between the unaltered glass and the alteration rim is sharp on a micrometer scale with no apparent chemical diffusion profiles. Such features are not compatible with a process that is solely controlled by diffusion of cations and hydrogen species. Here we suggest that during the first step the borosilicate glass dissolved congruently followed immediately by the precipitation of amorphous SiO<sub>2</sub> at an inward moving interface. Once the SiO<sub>2</sub> layer became too thick, the communication between fluid and the reaction interface ceased and the SiO<sub>2</sub> layer increasingly incorporated Ce, Ti, and Al. In a second step the Liesegang bands were formed by a diffusion-controlled process.

## Metamorphic reworking of the Congo Craton in Uganda

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The Basement Complex of Uganda (BC) forms the NE corner of the Archean Congo craton bordering the Pan-African Central African Fold Belt (CAFB) in the N and the East African Orogen (EAO) in the E. We studied the tectonic history of the Archean basement and of its Neoproterozoic cover series by means of metamorphic P-T paths and metamorphic ages (SHRIMP dating of zircon and U-Th-total Pb chemical dating of monazite).

A clockwise P-T evolution during a first metamorphism (M1) along the northern craton border is constrained by prograde sillimanite and later formed kyanite included in garnet, peak-metamorphic Grt-Sil-Bt assemblages and the late-stage formation of cordierite rims around garnet. Peak conditions (9-10 kbar; ca. 940 °C) were followed by decompression to about 8 kbar and subsequent near-isobaric cooling. A second metamorphism (ca. 750°C at 7 kbar) is evident from new garnet overgrowing Bt-Sil/Ky-Crd symplectites formed during retrogression after M1. Dating revealed ca. 2.6 Ga for M1 and ca. 650 Ma for the reworking, indicating that the granulites formed the deep cratonic crust for about 2 Ga prior to their Pan-African uplift.

At the eastern craton border, aluminous Opx (up to 11.5 wt% Al<sub>2</sub>O<sub>3</sub> when coexisting with Grt) and assemblages with Spl-Spr-Qtz-Grt-Osumilite (pseudomorph), Opx-Sil-Grt or inverted pigeonite point to extreme metamorphic temperatures (>1000°C) at 8-10 kbar in a very large area (including the Labwor Hills). In contrast, late-stage Grt-Cpx-Qtz coronas in charnockitic and metabasic rocks were formed at 6-7.6 kbar only after near-isobaric cooling to temperatures of 650-680°C. The reaction history in metapelites points to an anti-clockwise P-T evolution. UHT metamorphism and magmatism of high-temperature A type granitoids are spatially and timely associated and lasted for about 70 Ma between 700 and 630 Ma. The intrusion of the magmas is interpreted as the cause of the UHT metamorphism.

Neoproterozoic cover series (NW Uganda) have not experienced granulite facies metamorphism unlike the basement rocks on which they are resting. They are Grt-Ky-Ms-Bt-Pl-Qtz bearing micaschists metamorphosed at 8 kbar and 650-680°C during a clockwise P-T path. Monazites show a one-stage growth history and give ages of 621±26 and 633±27 Ma for the metamorphic event that was caused by thickening of the cratonic crust.