Electron-beam annealing in radiation-damaged zircon

Váczi, T.1,2,*, Nasdala, L.3

1 Wigner Research Centre for Physics, Hungarian Academy of Sciences, Budapest, Hungary
2 Department of Mineralogy, Eötvös Loránd University, Budapest, Hungary
3 Institut für Mineralogie und Kristallographie, Universität Wien, Vienna, Austria
* E-Mail: vaczitamas@caesar.elte.hu

It is known that heating to high temperatures is able to induce ordering (called thermal annealing) in damaged zircon (e.g. Geisler et al. 2001). It has been demonstrated that an electron beam is also effective in rearranging displaced atoms and increasing crystallinity (Váczi and Nasdala 2017). This contribution presents recent findings and an assessment of the real structure of minerals self-irradiated by alpha decay.

Actinide-containing minerals cause radiation damage in themselves (self-irradiation) or in their neighbouring phases (radiation haloes). Self-irradiation occurs mostly through the recoil of heavy daughter nuclei produced in alpha decay events, which gradually destroys the crystallinity of the host. Zircon, the most important accessory mineral for age determination, is notable for accumulating and preserving self-irradiation damage, the degree of which depends on crystallisation age, actinide content and thermal history.

Zircon may be heated to high temperatures inducing the recovery of damage through geological processes and in the laboratory. However, due to the importance of this mineral in geological investigations, electron-beam analysis is very commonly done. Recently, in a systematic study of a set of naturally damaged zircon samples (Váczi and Nasdala 2017) spanning a large range of radiation damage, it has been shown that an electron beam induces a dose-dependent, partial recovery of ion-irradiation damage. The recovery was observable through changes in Raman spectra recorded at electron probe analysis spots (Fig. 1).

![Raman map from the surface of zircon sample M144 (low to moderate initial damage) after electron irradiation. The “hills” reveal increased crystalline order at electron-beam impact spots, while the crosses show the guide marks milled using a focused Ga-ion beam.](image-url)
Our experimental observations and calculations revealed that electron-beam annealing is athermal. A rate change was also observed: following an initial rapid recovery of Raman band widths, the process becomes gradually slower (Fig. 2), and this pattern was recognisable in all samples. The decrease of the Raman band width is directly related to the initial damage level (in the case of small initial damage we saw little change in Raman widths). Using arguments based on a geometric model of recoil damage accumulation (Ketcham et al. 2013), we tentatively inferred that the observed recovery is actually larger in terms of phonon coherence length in the samples with less initial damage. Further study is in progress to clarify this in more detail.

Fig. 2. Damage evolution during electron-beam bombardment in sample G4 (heavy initial damage) as a function of irradiation duration and beam current

Based on experimental findings, the best practice during a multi-method analysis of zircon (and perhaps other accessory minerals as well) to characterise samples first by visible-light spectroscopy (Raman, photoluminescence etc.) and only subsequently turn to electron-beam methods. Our results show that short beam dwell times produce no observable spectroscopic changes (i.e., no rearrangement of ions), while prolonged exposures to the electron beam induce ion mobility in the irradiated volume. This should be kept in mind when planning the analytical agenda.

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References: