

Disturbance of the monazite U-Th-Pb chronometer by fluids: A study combining stable isotopes and *in situ* dating

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The monazite U-Th-Pb chronometer is extensively used to date metamorphic rocks because it is usually considered to be resistant to diffusive Pb loss at temperatures as high as 800°C. However, age perturbations are sometimes documented in monazite grains showing distinct compositional and age domains. The mechanism by which Pb is lost is not well understood. Fluid-assisted recrystallization has been shown to constitute an efficient mean to reset the monazite chronometer in experimental conditions (Seydoux-Guillaume *et al.* 2002). Whether the same holds true in natural systems remains to be documented.

Here we present *in situ* U-Th-Pb ages on monazite from foliated pegmatitic veins from highly strained levels of the Central Rhodope metamorphic pile (Greece, Bulgaria), together with oxygen isotope characterization and ⁴⁰Ar-³⁹Ar data. U-Th-Pb ages were obtained by (LA)-ICPMS on 8 monazite grains from three samples. In one sample from Bulgaria, the ages are concordant at 35 Ma, and no chemical zonation is observed. Quartz and feldspar have δ¹⁸O values consistent with equilibration at high temperatures. Microfabrics in the vein and the host rocks indicate high grade conditions of deformation. In the two other samples, from Greece, monazite grains show distinct chemical domains and yield discordant U-Th-Pb ages ranging from 52 to 36 Ma. Strong oxygen isotope disequilibrium between quartz and feldspar argues for a complex fluid history: high-δ¹⁸O fluids first invaded rocks and were followed by low-δ¹⁸O fluids at lower temperatures. Low temperature fluid circulation at around 36 Ma is consistent with ⁴⁰Ar-³⁹Ar dating of both samples at 33-34 Ma (mica plateau ages) and with microfabrics documenting a transition from high grade to greenschist facies conditions of deformation during intense shearing. These data confirm the capacity of fluids in disturbing significantly the monazite chronometer.

References

Seydoux-Guillaume A.M., Paquette J.L., Wiedenbeck M., Montel J.M. and Heinrich W. (2002). Experimental resetting of the U-Th-Pb system in monazite. *Chemical Geology* **191**, 165-181

Uranium isotope ratios determined by MC-ICP-MS and the assessment of total combined uncertainty budgets

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Precise determination of uranium isotopic composition is required in nuclear environmental and geologic studies. Particular applications imply determination of extremely low ²³⁶U quantities in the presence of a large excess of ²³⁸U and ²³⁵U which represent severe sources of interference in determinations of ²³⁶U.

A multi collector inductively coupled plasma mass spectrometer (MC-ICP-MS) - Nu Plasma HR - equipped with an ion deceleration filter was applied for rapid and sensitive determination of ²³⁴U/²³⁸U, ²³⁵U/²³⁸U and ²³⁶U/²³⁸U isotope ratios in aqueous samples on a concentration level typical for digested environmental samples. Application of the ion deceleration lens system in Nu Plasma ICP-MS improved significantly the abundance sensitivity and made possible the analysis of ²³⁶U/²³⁸U isotope ratios in the 10⁻⁸ to 10⁻⁷ range. On the other hand, using the ion deceleration lens for determination of ²³⁶U/²³⁸U isotope ratios above 10⁻⁵ was found disadvantageous because it might introduce additional uncertainty. Accuracy of uranium isotope ratio measurements by MC-ICP-MS was evaluated by analysis of unknown samples obtained in the frame of a round robin exercise as well as by comparative measurements using sector-field ICP-MS with single ion detector and thermal ionization mass spectrometry (TIMS). Combined uncertainty of measured ratios ranged from 0.17% to 0.20% for ²³⁵U/²³⁸U, from 1.0% to 1.1% for ²³⁴U/²³⁸U and from 1.3% to 53% for ²³⁶U/²³⁸U. The main uncertainty contribution component to the ²³⁵U/²³⁸U ratio measured by MC-ICP-MS is the uncertainty of the applied isotopic standard. The uncertainty budget for ²³⁶U/²³⁸U strongly depends on the abundance of ²³⁶U. In particular, peak tailing and interference by ²³⁵U¹H⁺ ions increased the measurement uncertainty in the case of ²³⁶U/²³⁸U lower than 10⁻⁷. Nevertheless, accurate determination of ²³⁶U/²³⁸U isotope ratios as low as 3x10⁻⁸ was possible, which opens new possibilities for tracing this isotopes in environmental samples by using MC-ICP-MS, in particular with respect to its better time-effectiveness and lower labor expenses when compared to other highly sensitive mass spectrometric methods.