

## Si isotope fractionation during silica precipitation in batch-reactor experiments

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Recent research has demonstrated the versatility of silicon isotopes in reconstructing pathways and processes in (bio)geochemical systems. Interpretations of data from siliceous chemical sediments in the rock record rely on quantitative insight into isotopic fractionation during precipitation from silica-carrying solutions. We performed batch-reactor experiments to explore the isotopic fractionation behavior of precipitating silica in low-temperature environments.

Supersaturated silica solutions were obtained by cooling a 90°C saturated solution to temperatures of 10-35°C in silica-seeded batch reactors. Decreased silica solubility induced by the temperature drop resulted in deposition of monomeric silica on the seeds. Solution samples, taken at regular time intervals, were analyzed for silicon isotopic composition by MC-ICP-MS with a typical precision of 0.12‰ (2σ) [1]. The isotope fractionation followed Rayleigh distillation trends and proceeded in two consecutive stages. Initially (stage I), at high degrees of oversaturation, fractionation factors ( $\epsilon_{\text{ppt-dis}}$ ) were negative and ranged between -1.1 and -2.0‰. Subsequently (stage II), when saturation levels had decreased, fractionation factors were positive and ranged from +0.4 to +0.7‰. The magnitude of the fractionation factor appeared to increase with decreasing temperature in both stages.

We propose that silicon isotope fractionation is controlled by the degree of oversaturation in the reactor, resulting in dominant precipitation in stage I and partial re-dissolution of precipitated silica in stage II. We hypothesize that fractionation during precipitation occurs due to energetically preferred breaking of <sup>28</sup>Si-O bonds in the dissolved silica ( $\epsilon < 0$ ), before formation of new Si-O-Si bonds at the surface of silica grains. In stage II, the system approaches equilibrium, and decreased oversaturation allows partial re-dissolution of Si from the surface with a relatively high rate for <sup>28</sup>Si. Consequently, more <sup>28</sup>Si goes back into the solution than precipitates, with the net effect that <sup>30</sup>Si is preferentially retained in the solid ( $\epsilon > 0$ ).

Our results suggest that the ultimate isotopic composition of cherts and non-biogenic silica deposits is a function of the degree of oversaturation, for example in cases where rising hydrothermal fluids with different loads of dissolved silica cool down to the same ambient temperature when venting at the seafloor.

[1] Van den Boorn, S.H.J.M. *et al.* (2006) *J. Anal. At. Spectrom.* **21**, 734-742

## Discordant polymetamorphic zircons: the rule in crystalline nappes of the SW Norwegian Caledonides

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### 5a. What zircons tell us about crustal evolution

Resetting of zircons is commonly due to Pb loss related to metamictisation and subsequent alteration, or to other processes such as strain and recrystallization disturbing the crystal lattice. In general it is possible to identify domains of zircons that have escaped partial resetting. Such domains can then be isolated and analyzed separately. In the crystalline nappes of the SW Norwegian Caledonides, however, zircon populations typically yield data that are variously discordant, defining either discordia lines or, in more complex cases, scattered arrays. The most common pattern is exemplified by zircons in orthogneisses from a klippe on Hardangerjøkulen near Finse, that defines a line with an upper intercept age of  $1658 \pm 9$  Ma, indicating magmatic formation, and a lower intercept age of  $968 \pm 26$  Ma, reflecting the Sveconorwegian metamorphic overprint. The discordia line is defined by seven precise ID-TIMS measurements on clear, inclusion free, optically undisturbed zircon fractions on mainly short prismatic grains subjected to either air or chemical abrasion. The fact that even chemical abrasion did not remove the discordance indicates that the discordance is frozen in by recrystallization. The grains themselves show little indication of metamorphic new growth. Coexisting titanite, however, formed during Sveconorwegian metamorphism. Although the nappe was emplaced during the Caledonian orogeny, the U-Pb data were left nearly completely unaffected by these secondary events. This type of behavior is very common in the gneisses of comparable nappes in the Hardanger-Ryfylke Complex to the south and the Jotun Complex to the north of Hardangerjøkulen, although a Caledonian influence is more evident in some cases and creates more scatter. A very different U-Pb discordant pattern is observed instead in anorthositic-jotunitic gneisses of the northern Lindås Nappe, where the Caledonian amphibolite facies overprint and local deformation strongly affected the zircon populations, causing strong Caledonian resetting and local new growth. In zircons of one of the anorthosite localities the resetting can be linked to strain and plastic deformation, but the other key factors seem to be metamorphic reactions and the release of chemicals that allow to form new zircons.