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## The effect of thermochemical equalization of the isotopic compositions of U (IV) and U (VI) during the recrystallization of metamict structures

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It is well known that metamict minerals are regarded as natural analogues of aged HLW forms. Given that nuclear decay can cause significant heating of the corresponding waste forms, it is certainly of interest to take a close look at the thermochemical processes in these minerals. It has been shown that the <sup>238</sup>U and <sup>234</sup>U isotopes exhibit non-identical thermochemical behavior in the course of isochronal annealing up to 600 °C, that is, until the metamict structure begins to recrystallize. It has also been observed that, as the crystal structure forms in britholite, the isotopic compositions of the U (IV) and U (VI) valence forms equalize. It is, therefore, more than relevant to find out to what extent this equalizing effect is characteristic of other metamict minerals. The test object in this study was a metamict polycrase-(Y) with insignificant inclusions (according to an XRD-analysis) of fergusonite-(Y) and hydroxybetafite. EMP- and XRD-analyses showed that the elemental composition of the sample corresponds to the formula:  $(Y_{0.37}Ca_{0.17}U_{0.05}Ag_{0.05})_{(0.64)}(Ti_{0.71}Nb_{0.58}Si_{0.42}Fe_{0.23}Ta_{0.06})_{(2.00)}(O_{4.11}OH_{1.89})_{(6)}$ . Thermal annealing of different samples was carried out in a vacuum for ten minutes at three different temperatures. Separation of the valence forms of uranium and alpha spectrometric analysis were performed according to the method used in. The table below provides the activity ratios – AR (<sup>234</sup>U / <sup>238</sup>U) – for the total uranium U (Σ) and for the valence forms U (IV) and U(VI). The AR (Σ) value, equal to 0.96, shows that the initial sample is characterized by a deficiency of the radiogenic uranium isotope. It should be noted that all the uranium is oxidized to the hexavalent state. Heating the sample to 200°C leads to the reduction of a small fraction of the uranium (about 14-15 percent). At 600°C, the reduction process continues, and the share of U (IV) is close to 31-32 percent. When the metamict structure is fully crystallized at 800°C, the values of AR (<sup>234</sup>U/<sup>238</sup>U) in both valence forms equalizes and acquires the initial value AR (Σ)=0.96. The very same non-identical behavior of the uranium isotopes in the course of the annealing process and the equalization of AR (IV) and AR (VI) values during the crystallization of the metamict structure have been discovered in experiments with britholite.

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