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A revised crystal structure for the rare earth fluoride gagarinite-(Ce) from experimental synthesis by fluid-induced alteration of chevkinite-(Ce)

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The Rare Earth Element (REE) fluoride, gagarinite-(Ce), ${}^A\text{Na}^B(\text{REE,Ca})_2\text{F}_6$ has been synthesized in an experiment designed to examine the fluid-induced alteration of chevkinite-(Ce). The experiments were conducted at 600°C and 400 MPa for 21 days, 550°C and 200 MPa for 63 days and 600°C and 200 MPa for 42 days. The formula of crystallized gagarinite-(Ce) analogue is $(\text{Na}_{1.10}\text{Ce}_{0.69}\text{Ca}_{0.44}\text{Nd}_{0.31}\text{La}_{0.26}\text{Pr}_{0.12}\text{Sm}_{0.04}\text{Sr}_{0.03})_{3.0}\text{F}_{6.0}$.

The mineral, previously named zajacite-(Ce), is known from only one natural occurrence, a hypersolvus granite from the Strange Lake Zr-Y-REE-Nb-Be deposit, Quebec-Labrador. The space group was identified as $P\bar{3}$ (Jambor et al. 1996) and later $P6_3/m$ (Sciberras et al. 2011).

The gagarinite-(Ce) in our experiments crystallized in $P\bar{6}$, with $a = 6.1465(2)$, $c = 3.75950(10)$, $R_1 = 1.37\%$. We observed 26% twinning by a twin centre.

Previous studies assumed full occupancy of the A site. To charge balance the substitution $2{}^A\text{REE}^{3+} \rightarrow {}^A\text{Ca}^{2+} + {}^A\text{REE}^{3+} + {}^B\text{Na}^+$, the amount of extra Na^+ must equal Ca^{2+} in the final formula, giving $\text{Na}_x(\text{Ca}_x\text{REE}_{2-x})\text{F}_6$. Gagarinite-(Ce) from the experiment shows surplus Na over Ca with a ratio close to 2:1. A vacancy in the REE site is necessary, equal to 1/3 of the redundant Na to remain in charge balance. If the Na content exceeds 1 in the formula unit, Na has to substitute for REE in the cation site. Thus, the overall substitution mechanism is:

$(2x-y)\text{REE}^{3+} \rightarrow y {}^B\text{Na}^+ + (x-y) {}^A\text{Na}^+ + y {}^A\text{Ca}^{2+} + (x-y)/3 {}^A\Box$, where \Box stands for vacancy and $x > y$.

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References

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