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Theoretical modeling of high uranium zircon's crystal morphology according to atomistic calculation's data

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The unique properties of zircon (resistance to weathering, various isotope-geochemical heterogeneities, etc.) are commonly used for geochronology and reconstruction geological process. Moreover, zircon is highly resistant to radioactive exposure. It is of great interest in solving both fundamental and applied problems associated with the isolation of high-level radioactive waste.
The atomistic calculations of crystal surfaces are commonly performed using the energetic characteristics of faces, namely, the surface energy Esurf, which is a measure of the thermodynamic stability of the crystal face.
As we know from our previous research (Gromalova et al., 2012) depending on the content and distribution of the impurity over the positions of the mineral structure occur a change in its morphology of crystals. Furthermore one of the constitutive factors affecting the value of the Esurf in calculations is a choice of potentials model (Gromalova et al, 2015). So, on the basis of the previously optimized potential model (Gromalova et al.,2017), we carried out an atomistic simulation of morphology of high-uranium zircon's crystals.
The surface energies for different faces of zircon were calculated using Metadise code (Watson et al., 1996). The computation time for one simple form was from 30 minutes to 12 hours.
Zircon crystal structure has 4 non-equivalent zirconium positions. The Zr coordinates x,y,z for 1-4 positions are 0.000,0.750,0.125; 0.000,0.250,0.875; 0.500,0.750,0.375; 0.500,0.250,0.675, respectively. Substitution of zirconium for uranium was carried out in a standard cell (I41/amd) for one or two (in various combinations) positions.
Calculations showed that an increase of the uranium impurity content in zirconium position (from 25 to 50 %) leads to a changing of number of faces, namely, to signification loss of the morphological significance of the zircon prism (110), until it disappears completely in the case of uranium substitution of the zirconium 1-4 and 2-3, flattening of the habit, disappearance of the face (101), and an increase in the morphological significance of bipyramid (111). In the case of substitution of U positions 2-4 of Zr, morphological significance of bipyramid (111) and prism (100) are practically equal.
In general, it is possible to observe depletion of zircon's shape in the case of an increase content of uranium impurity, which agrees well with the observations (Shiryaev et al., 2016) of chernobylite. The research showed that mineral faces depends not only on external parameters, but also depends of atomistic filling, and especial of impurites, that make up the structure.

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