Thorium-rich intergrowns and inclusions in monazite-(Ce) samples with low and high actinide content

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Monazite, although frequently contains significant amount of thorium and uranium, never exhibits the properties of metamict minerals. Routine X-ray diffraction analysis yields typical X-ray diffraction pattern of monazite showing no indication of metamictization. This is attributed to its crystal structure properties, especially to the P-O bonds which are shorter and stronger when compared to Si-O bonds in zircon, promoting resistance to radiation damage [1]. Critical temperature of amorphization ranges between 150 and 200°C enabling an early healing of the radiation damage in the crystal structure [2].

Two monazite-(Ce) samples were studied and compared for their crystal-chemical properties: monazite-(Ce) from Eptevann, Norway, originates from a granitic pegmatite while monazite-(Ce) from Eureka Farm, Namibia, is related to carbonatite deposits

The two samples were investigated by X-ray diffraction (XRD) and scanning electron microscopy (SEM) using Tescan TS5136MM SEM with energy dispersive system (EDS) mounted to the instrument (Oxford Inca system). The samples were subjected to annealing experiments in order to monitor the changes in crystallite size and strain parameters.

Monazite-(Ce) from Eptevann contains more than 10% of actinides on oxide basis, mostly as ThO₂. However, the mineral does not exert physical properties of a metamict mineral as well as X-ray data show characteristic monazite diffraction pattern yielding unit cell parameters as follows: a = 6.473(1) Å, b = 6.986(1) Å, c = 8.178(1) Å, $\beta = 126.511(8)^{\circ}$. After annealing experiments at 200, 500 and 1000°C unit cell parameters decrease, the intensity of diffraction maxima increases whereas their width decreases as the temperature rises. The calculated crystallite size and strain show a decrease in strain and increase in crystallite size with temperature increase. Thorium, although being significantly incorporated in the structure of monazite, can be also found in a phase intergrown with monazite (Figure 1). ED spectra show that the intergrown phase is a modification of ThSiO₄, most likely huttonite. The occurrence of the phase is rather irregular showing no crystal form.

X-ray diffraction pattern of monazite-(Ce) from Eureka Farm indicates no other phases beside monazite, yielding monazite unit cell as follows: a = 6.4987(3) Å, b = 7.0459(3) Å, c = 8.2478(3) Å, $\beta = 126.486(2)^{\circ}$. Crystallite size and strain parameters do not change significantly during annealing experiments with a starting crystallite size being larger and a starting strain being smaller when compared to the monazite sample from Eptevann. This can be related to its very low actinide content, as indicated by EDS. The BSE image clearly shows the occurrence of mineral inclusions in thermally untreated monazite (Figure 2). Chemical composition unambiguously shows the presence of thorianite (ThO₂) what is also indicated by its almost regular cubic morphology. It is more challenging to identify another phase which might be assigned as a silicate containing significant amount of thorium and iron.

The occurrence of different thorium-rich phases in monazite-(Ce) samples with different actinide content could be related to their different origin and thermal history. The intergrowns in monazite from Eptevann can be considered to be exsolutions from Th-rich regions in crystallizing monazite, with both monazite and huttonite sharing the same crystal structure type. On the other hand, inclusions in monazite from Eureka Farm are more likely micro-phases trapped in monazite during crystallization.

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Figure 1. BSE image of monazite-(Ce) from Eptevann, showing exsolutions of thorium silicate, most likely huttonite



Figure 2. BSE image of monazite-(Ce) from Eureka Farm, showing the inclusion of thorianite (ThO_2) and another Fe-Th-reach mineral phase