Phase Separation in Fluorite-Related U 1– y Ce y O 2– x : A Re-Examination by X-ray and Neutron Diffraction

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The phase separation observed at low temperature (below circa 600 K) in the U1-yCe y O2-x system and for values of y between roughly 0.34 and 0.5 purportedly involves fluorite structures only [12,3]. All these studies were therefore performed using X-ray diffraction techniques and then are not able to track evolution of the oxygen-sub lattice. In this work, the phase separation in U0.54Ce0.46O2-x has been reexamined using Xray and neutron diffraction. As neutrons' scattering lengths for U and O do not differ t0o much, information about the oxygen sub lattice can be obtained from neutron diffraction patterns. Below a critical temperature, the existence of two fluorite related structures in the miscibility gap is confirmed: a stoichiometric U0.54Ce0.46O2 phase and an oxygen-deficient U0.54Ce0.46O2x phase. Although the former is indeed a fluorite, we show that the other end-member phase has a C-type bixbyite structure. This would suggest that the oxygen-deficient phase can be described as a bixbyite over the entire cerium composition range (see figure 1). In this work, we present some recent developments performed to understand the phase transformation in this material and try to rely this with the defects produced by the oxygen deficiency in the anionic sub lattice. The ultimate goal of this approach is to simulate this phase transformation within the Phase Field approaches of phase transition.

Figure 1 : Schematic description of two ended phases produced during the phase transformation of (U,Ce)O1-x (x=0.07).

References:

[1] Markin, T.; Street, R.; Crouch, E. The uranium-cerium-oxygen ternary phase diagram. J. Inorg. Nucl. Chem. 1970, 32, 59–75.

[2] Lorenzelli, R.; Touzelin, B. Sur le système UO2-CeO2; etude cristallographique àhaute température. J. Nucl. Mater. 1980, 95, 290–302.

[3] Vauchy, R.; Belin, R. C.; Richaud, J.-C.; Valenza, P. J.; Adenot, F.;Valot, C. Studying radiotoxic materials by high temperature X-raydiffraction. Applied Materials Today 2016, 3, 87–95.

