

## Addition of Cerium and Cesium oxides to the $B_2O_3$ - $FeO_x$ - $UO_x$ - $ZrO_2$ database

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The oxide system  $Cs_2O$ - $CeO_x$ - $B_2O_3$ - $FeO_x$ - $UO_x$ - $ZrO_2$  is relevant for the melting behavior of nuclear fuels at high temperatures, Cerium and Cesium oxides being present in radioactive and toxic wastes. In case of nuclear accidents, such as the Fukushima accident, molten nuclear fuels based on  $UO_2$ - $ZrO_2$  system will react with various kinds of surrounding materials in fuel container (e.g. Fe in stainless steel) to form multicomponent debris. A thermodynamic database for the entire system was so far not available, however a thermodynamic description of the  $B_2O_3$ - $FeO_x$ - $UO_x$ - $ZrO_2$  is a part of the TDNucl database [TDNucl] whereas the Cerium and Cesium oxides were not present. The additional new 9 quasi-binary and 3 ternary systems containing Cerium and Cesium oxides have been thermodynamically assessed using all presently available experimental data thus completing the required oxide database.

The Gibbs energy of the liquid phase has been modelled using a non-ideal associate solution model. The compositions of the pure liquid oxide species as well as the associates have been chosen to have two moles of cations per associate thus keeping the successful method of Spear and Besmann [Bes2002].

In the thermodynamic assessments of the binary systems  $B_2O_3$ - $Cs_2O$ ,  $Cs_2O$ - $FeO$ ,  $Cs_2O$ - $Fe_2O_3$ ,  $Cs_2O$ - $UO_3$ ,  $Cs_2O$ - $ZrO_2$ ,  $CeO_2$ - $FeO$ ,  $Ce_2O_3$ - $Fe_2O_3$ ,  $CeO_2$ - $UO_2$ ,  $CeO_2$ - $ZrO_2$ ,  $Ce_2O_3$ - $ZrO_2$  as well as the ternary  $Cs$ - $Fe$ - $O$ ,  $Cs$ - $O$ - $U$ ,  $Ce$ - $Fe$ - $O$  systems 25 stoichiometric solid phases and 4 solid solution phases were incorporated using available experimental information.

The thermodynamic description of the stoichiometric phase Cerium Zirconat  $Ce_2Zr_2O_7$  was assessed taking into account the experimental determined by Bolech [97Bolech] heat capacity and thermal stability given by Leonov, Keler and Andreeva [96Leonov].

The ternary  $CeO_2$ - $ZrO_2$ - $UO_2$  system is characterized by complete solubility between the corresponding oxides at high temperatures. The phase  $MeO_2$ -HT was modelled using the following formula  $(Ce^{+3}, Ce^{+4}, U^{+2}, U^{+4}, Zr^{+2}, Zr^{+4})(O^{-2}, Va)_2$  which allows to describe the mutual solubility of the zirconium, uranium and cerium oxides.

Particular attention was also given to the phases  $ZrO_2$ -MT and  $ZrO_2$ -LT which exhibit wide solubility with respect to cerium oxide.

### Literature

- [TDNucl] Thermodata Nuclear phase diagrams, 2012,  
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