Thermodynamic modelling of U-O-X systems with X=Pu,Zr,C - Application to nuclear fuel materials studies

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Thermodynamic data and phase diagrams on actinide multi-component systems are necessary to assess in order to predict the chemical behaviour of the nuclear fuel materials at high temperature in reactor in normal and off-normal conditions. Since 2005 the FUELBASE thermodynamic database is developed using the CALPHAD method to provide a flexible tool to perform thermodynamic calculations on advanced nuclear fuel materials for the Sodium Fast Reactor, the Gas-cooled Fast Reactor and the High Temperature Reactor in the frame of ACTINET network and F-BRIDGE European project. The thermodynamic modelling of the oxide fuels UO_2 and $(U,Pu)O_2$ is a challenge considering the complexity of both the variation of the thermochemical properties with the departure from stoichiometry (oxygen/metal ratio) and temperature as well as the oxide rich part of phase diagrams. A three sublattice model with ionic species $(U^{3+},U^{4+},U^{5+},Pu^{3+},Pu^{4+},Zr^{2+},Zr^{4+})_1(O^{2-},Va)_2(O^{2-},Va)_1$ is developed to describe the fluorite phase where the first sublattice corresponds to the site for the metallic cations, the second one is the normal site for oxygen anions and the third one is the site for interstitial oxygen anions. Oxygen vacancies and interstitials are introduced on the second and third sublattices to describe respectively the hypostoichiometric and hyperstoichiometric regions of the fuel. The ionic two-sublattice model is used to describe the liquid phase. Some applications of the thermodynamic calculations will be given. Calculations of oxygen potential data of MOX fuels will be presented using the thermodynamic description of the U-Pu-O system. Calculated phase diagrams will be also presented to show the composition range of the fuel where decompositions phenomena can occur. Another application will be related to the interaction between the fuel UO_2 and the Zr cladding during a severe nuclear accident using the database on the U-O-Zr system. Finally, calculations of CO and CO_2 equilibrium pressures resulting from the interaction between the UO_2 fuel and the carbon buffer in TRISO particles will be presented.