

Thermochemical modeling of advanced oxide and carbide nuclear fuels within the FUELBASE project

C. Guéneau ^a, A. Berche ^a, S. Chatain ^a, J.C. Dumas ^b, C. Martial ^b, P. Gotcu ^c, O. Benes ^a, N. Dupin ^d, B. Sundman ^e, C. Chatillon ^f, T.M. Besmann ^g, R.J.M. Konings ^c

^a DEN/DANS/DPC/SCP/LM2T – CEA Saclay – 91191 Gif-sur-Yvette cedex – France

^b DEN/CAD/DEC/SESC/LLCC – CEA Cadarache – 13108 Saint-Paul Lez Durance
Cedex – France

^c Joint Research Institute for Transuranium Elements – 76125 Karlsruhe – Germany

^d Calcul Thermo – 63670 Orcet – France

^e INSTN – CEA Saclay – 91191 Gif-sur-Yvette cedex - France

^f CNRS / SIMAP – 38402 Saint-Martin d'Hères Cedex – France

^g Oak Ridge Nat. Lab. – Oak Ridge, TN 37831-6063 - US

Mixed oxide and carbide of uranium and plutonium are considered as reference fuels for respectively the Sodium Fast Reactor and the Gas Fast Reactor. Oxide fuels (MOX) have been extensively investigated in the past. Nevertheless some uncertainties remain on the phase diagram of the U-Pu-O system at low temperature where the MOX fuels containing high Pu content can decompose into two different phases for O/metal ratio lower than 2 and at high temperature where the solidus and liquidus temperature are still not well known. Moreover, the effect of the addition of the minor actinides Am, Np and Cm on both phase diagrams and thermodynamic properties such as oxygen chemical potential has to be investigated. In case of the carbide fuels, thermodynamic properties and phase diagram of the U-Pu-C system have been mainly investigated in the 60-70's. As for the oxides, some uncertainties remain on phase diagrams and on some thermodynamic properties. For example, the relative stabilities of the different uranium carbides U_2C_3 and UC_2 as a function of the temperature is still a subject of controversy and requires new determinations. Some data are missing on the Pu-C system. Within the European ACTINET network and F-BRIDGE project, we develop the thermodynamic database FUELBASE to provide a flexible tool to perform thermodynamic calculations on these advanced fuels in parallel to the experimental work. The FUELBASE database is developed using the CALPHAD method. The first step consists in a critical review of all the existing experimental data on phase diagrams and thermodynamic properties. Some models are developed to describe the variation of the Gibbs energy of the phases (liquid, solid and gas) as a function of temperature and composition. The Gibbs energy functions are derived from a least-square minimization method using the available experimental data. A thermochemical model is available for both oxide and carbide fuels. The addition of the minor actinides to the U-Pu-O system is in progress.