THERMOPHYSICAL AND CHEMICAL PROPERTIES OF MINOR ACTINIDE FUELS

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Abstract

As the world's nuclear power programme evolved, much theoretical and experimental effort was applied to studying the basic properties of the fuels: first uranium dioxide and then mixed uranium – plutonium dioxide. Recommendations were produced for all the relevant physical properties of UO_{2+x}, as a function of temperature up to and beyond the melting point, and also as functions of burnup and of the oxygen-to-metal ratio (O/M, often described by the deviation x from stoichiometry). Experiments were also carried out to determine many of the properties of the mixed oxide $U_{1-y}Pu_yO_{2+x}$ (where x can be negative as well as positive), which typically also vary with the plutonium fraction y. Where data on the effect of Pu fraction were unavailable, the uranium dioxide properties were often deemed to be a good enough approximation for properties of the mixed oxide. Less effort was applied to the carbide and nitride fuels. Good data exist for the unirradiated materials, but because of the limited irradiation experience of these fuels compared to oxide, evolution with burnup is less well understood.

The properties of advanced fuels proposed for minor actinide burning are much less well-known. A generalised oxide fuel could be $(U,Pu,Am,Np,Cm)O_{2+x}$; a uranium-free fuel with a diluent or inert matrix might be $(Pu,Am,Np,Cm,Zr)O_{2+x}$. or $(Pu,Am,Np,Cm,Mg)O_{2+x}$. Three extra actinides plus possible diluents of ZrO_2, Y_2O_3 or CeO_2, or an MgO inert matrix, give seven more dimensions of parameter space to be explored in addition to the four existing ones of Pu fraction, O/M ratio, temperature and burnup. Moreover, the high gamma activity of Cm demands that experimental facilities are much more heavily shielded than those for the alpha emitters U and Pu. Not surprisingly, many of the physical and chemical properties of these advanced fuels presently have to be estimated or inferred.

With such a spectrum of possible fuels, modelling plays a vital part in selecting a candidate for future development. The modelling codes need these physical properties, cast in a usable form. This paper surveys critically the available data for thermophysical and chemical properties of minor-actinide and diluent oxides, and makes recommendations for fission energy, fission products, thermal expansion, density and lattice parameter, melting points, enthalpy and specific heat capacity, thermal conductivity and elastic constants. In several cases, the available data seem to be poor and judgements are required. Some data are missing: the most important missing items are highlighted. Often, the best approximation for missing data is to follow the established practice and extrapolate the UO_2 values. Finally, the paper presents an assessment of the phase relationships and thermodynamic parameters for the actinide – diluent oxide system.