

First Principles CANDU Fuel Model

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Extended Abstract:

Many modeling projects on nuclear fuel rest on a quantitative understanding of the co-existing phases at various stages of burnup. Since the various fission products have considerably different abilities to chemically associate with oxygen, and the O/M ratio is slowly changing as well, the chemical potential (generally expressed as an equivalent oxygen partial pressure) is a function of burnup. Concurrently, well recognized small fractions of new phases such as inert gas, noble metals, zirconates, etc. also develop. To further complicate matters, the dominant UO₂ fuel phase may be non-stoichiometric and most of minor phases have a variable composition dependent on temperature and possible contact with D₂O in the event of a sheathing defect.

A Fuel Thermodynamic Model (FTM) to predict the phases in partially burned CANDU nuclear fuel containing many major fission products has been developed [1-4]. This model is capable of handling non-stoichiometry in the UO₂ fluorite phase, dilute solution behaviour of significant solute oxides, noble metal inclusions, a second metal solid solution U(Pd-Rh-Ru), zirconate and uranate solutions as well as other minor solid phases, and volatile species within the inert gas (Xe) plenum.

The treatment is a melding of several thermodynamic modeling projects dealing with isolated aspects of this important multi-component system. To simplify the computations, the number of elements has been limited to twenty major representative fission products known to appear in spent fuel. The proportion of elements must be first generated using SCALES-5. Oxygen is inferred from the concentration of the other elements. Provision to study the disposition of very minor fission products is included within the general treatment but these are introduced only on an as needed basis for a particular purpose. The building blocks of the model are the standard Gibbs energies of formation of the many possible compounds expressed as a function of temperature. To these data are added mixing terms associated with the appearance of the component species in particular phases.

To validate the model, experiments relating the chemical potential of oxygen to the moles of oxygen introduced to SIMFUEL* are underway. Existing measurements for irradiated fuel, both defected and non-defected [5], are also being incorporated into the validation process.

* Mixed sintered oxides samples to represent fission products present at different burnups of nuclear fuel

A solid-state electrochemical technique is used to both detect the oxygen potential in an oxidizing gas mixture as well as to measure the moles of oxygen introduced into the sample by exposure to the gas mixture. The equipment as discussed by Verrall [6], operates by heating the sample to 1000°C and passing an Ar/ H₂/H₂O gas mixture over the samples. The oxygen pickup is detected by an increase in the H₂ concentration in the Ar gas mixture. The H₂ content in this gas is determined down stream using a ceramic zirconia/calcia electrochemical cell which measures the current associated with the passage of O²⁻ ions through the solid state electrolyte necessary to convert all the H₂ to H₂O. The integrated current is proportional to the moles of hydrogen oxidized in the solid-state electrochemical cell. The difference in H₂ concentration before and after passing over the fuel sample provides a measure of oxygen pickup. The same technique can be applied in the reducing sense when studying oxidized fuel associated with defective fuel discharged from a reactor.

The validated Fuel Thermochemical Model is expected to have many applications including new fuel design, spent fuel chemistry and in the case of defective fuel (cladding breach), safety issues such as coolant chemistry, fuel bay chemistry and long term storage. The treatment has all ready been applied as a boundary condition in kinetical models of defective fuel oxidation.

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- 6) R.A. Verrall, and J.F. Mouris, "Capability for Measurement of O/U Ratios in As-Discharged Defected CANDU Fuel". 7th International Conference on CANDU Fuel: Kingston, ON, (2005).