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M O N T E C U C C O L I N O
memorandum

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Actinide Dioxides lattice parameter

The simulation of nuclear systems by means of numerical codes, such as ERANOS[1] or MCNP[2], requires the compiling of proper input files, describing the geometric configuration and material composition of the reactors. The comparison of the results obtained with different codes (needed in order to design new systems, due to the different transport equation solution models implemented in such codes) may hold if and only if it is found to fulfill equivalence criteria which guarantee the mutual consistency of the simulations.

One of the most important parameters to be considered for the equivalence of the two different calculations is the mass of fissile in the fuel region. In particular, in case of Mixed OXide (MOX) fuel, besides the isotopic characterization of the vectors, the components fractions of the mixture is needed. Within the ERANOS input file, it is possible to define several "Materiaux" which make up a "Milieu" by specifying their volumetric fractions (VFs) in the fuel. On the other side, MCNP needs every cell to be filled with a single material, which composition can be specified by means of a "mat card" defined through either atomic or weight fractions (respectively #Fs and WFs) of the composing isotopes mixture.

It is therefore needed a relation for the conversion of the VF into either #F or WF, and vice versa. As an example, taking into account the #F at first, it can be noticed that the commonly used PuO_x VF, defined as

$$VF = \frac{V_{\mathrm{PuO_{x1}}}}{V_{\mathrm{PuO_{x1}}} + V_{\mathrm{UO_{x2}}}}\,, \tag{1}$$

can be rewritten to highlight the dependence on the #F as

$$VF = \frac{1}{1 + \frac{n_{\mathrm{UO_{x2}}}}{n_{\mathrm{PuO_{x1}}}} \frac{v_{\mathrm{UO_{x2}}}}{v_{\mathrm{PuO_{x1}}}}}\,, \tag{2}$$

where both the molecules number $n_{\mathrm{ActO_x}}$ and the molecular volumes $v_{\mathrm{ActO_x}}$ (see the following section for further details) have been introduced.

Similarly the #F can be written as

$$\#F = \frac{n_{\mathrm{PuO_{x1}}}}{n_{\mathrm{PuO_{x1}}} + n_{\mathrm{UO_{x2}}}} = \frac{1}{1 + \frac{n_{\mathrm{UO_{x2}}}}{n_{\mathrm{PuO_{x1}}}}}\,. \tag{3}$$

[1] G. Rimpault *et al.* Schema de Calcul de Reference du Formulaire Eranos et Orientations pour le Schema de Calcul de Projet. Technical Report XT-SBD-0001, CEA, August 1997.

[2] F. B. Brown *et al.* MCNP Version 5. *Trans. Am. Nucl. Soc.*, 87:273, 2002.

By retrieving the molecules numbers ratio from (2) and substituting into (3) it can be found the desired relation:

$$\#F = \frac{1}{1 + \frac{v_{\mathrm{PuO_{x1}}}}{v_{\mathrm{UO_{x2}}}} \frac{1-VF}{VF}} \,. \tag{4}$$

The WF can also be obtained similarly, by introducing into the latter expression the molecular masses $m_{\mathrm{ActO_x}}$:

$$WF = \frac{1}{1 + \frac{m_{\mathrm{UO_{x2}}}}{m_{\mathrm{PuO_{x1}}}} \frac{v_{\mathrm{PuO_{x1}}}}{v_{\mathrm{UO_{x2}}}} \frac{1-VF}{VF}} \,. \tag{5}$$

Lattice parameter

The key parameter for the conversion of VF into either #F or WF is the molecular volume $v_{\mathrm{ActO_x}}$. It can be retrieved by approximately considering the $\mathrm{ActO_x}$ lattice as a regular structure of cubes with both edge and pitch $\lambda_{\mathrm{ActO_x}}$, the sc *lattice parameter*.

Several evaluations of actinides dioxides lattice parameters can be found both in literature and in nuclear databases. Referring to the most recent and exhaustive paper[3], the main actinide dioxides lattice parameters are (at 298 K):

Dioxide	λ [Å]
UO_2	5.4704
PuO_2	5.3960

For nonstoichiometric molecules, the lattice parameter is found to be slightly modified from the reference values tabulated: in particular, different behaviors of the lattice parameter are found for different O contents (x).

For $\mathrm{UO_x}$ a decrease of the lattice parameter is observed for increasing x and vice versa. From experimental observations it could be possible to extrapolate the two relations[3,4]

$$a_0\,[\text{Å}] = 5.4704 + 0.0175\,(2 - \mathrm{x}) \tag{6a}$$

$$a_0\,[\text{Å}] = 5.4704 + 0.094\,(2 - \mathrm{x}) \tag{6b}$$

for $\mathrm{x} < 2$ and $\mathrm{x} > 2$ respectively.

[3] Yamashita *et al.* Thermal expansions of NpO_2 and some other actinide dioxides. *J. Nucl. Mat.*, 245:72–8, 1997.

[4] N. A. Javed. Thermodynamic study of hypostoichiometric Urania. *J. Nucl. Mat.*, 43:219–24, 1972.

On the other side, PuO_x exibits an opposite trend, with increasing lattice parameter for increasing O content, according to the relations[5,6]

$$a_0\left[\text{Å}\right] = 6.1503 - 0.3789\,\text{x} \tag{7a}$$

$$a_0\left[\text{Å}\right] = 5.3643 + 0.01746\,\text{x} + 0.004\,U\left(\text{x} - 2.025\right) \tag{7b}$$

fitting experimental data respectively for $\text{x} < 2$ and $\text{x} > 2$. The function $U(t)$ appearing in (7b) is the unitary step function, defined as

$$U(t) = \begin{cases} 0 & t < 0 \\ 1 & t \geq 0 \end{cases}$$

Temperature effect on lattice parameter

The effect of the thermic dilatation on the lattice structure of actinide dioxides has been deeply investigated in literature in order to retrieve a linear expansion law for the lattice parameter as a function of temperature.

In particular, for the main actinide dioxides (UO_2 and PuO_2), the following expressions are here considered:

- UO_2 (Fig. 1):
  - Martin[7]

$$L(T) = \begin{cases} L(273)\left(9.9734\cdot 10^{-01} + 9.802\cdot 10^{-06}\,T - 2.705\cdot 10^{-10}\,T^2 + 4.391\cdot 10^{-13}\,T^3\right) \\ \qquad 273K \leq T \leq 923K\,, \\ L(273)\left(9.9672E-01 + 1.179E-05\,T - 2.429E-09\,T^2 + 1.219E-12\,T^3\right) \\ \qquad 923K \leq T \leq 3120K\,; \end{cases} \tag{8}$$

  - Yamashita *et al.*[3]

$$L(T) = 5.45567\cdot 10^{-01} + 4.581\cdot 10^{-06}\,T + 1.0355\cdot 10^{-09}\,T^2 - 2.736\cdot 10^{-13}\,T^3\,. \tag{9}$$

- PuO_2 (Fig. 2):
  - Yamashita *et al.*[3]

$$L(T) = 5.38147\cdot 10^{-01} + 4.452\cdot 10^{-06}\,T + 7.184\cdot 10^{-10}\,T^2 - 1.995\cdot 10^{-14}\,T^3\,. \tag{10}$$

Such relations, obtained for stoichiometric dioxides, have been found to satisfy the dilatation behavior of nonstoichiometric molecules too.

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[5] L. A. Morales, J. M. Haschke and T. H. Allen. Kinetics of reaction between Plutonium dioxide and water at 25C to 350C: formation and properties of the PuO_{2+x} phase. Technical Report LA-13597-MS, LANL, May 1999.

[6] J. M. Haschke and T. H. Allen. Equilibrium and thermodynamic properties of the PuO_{2+x} solid solution. *J. Alloys Comp.*, 336:124–31, 2002.

[7] D. G. Martin. The thermal expansion of solid UO_2 and (U,Pu) mixed oxides - A review and recommendations. *J. Nucl. Mat.*, 152:94–101, 1988.

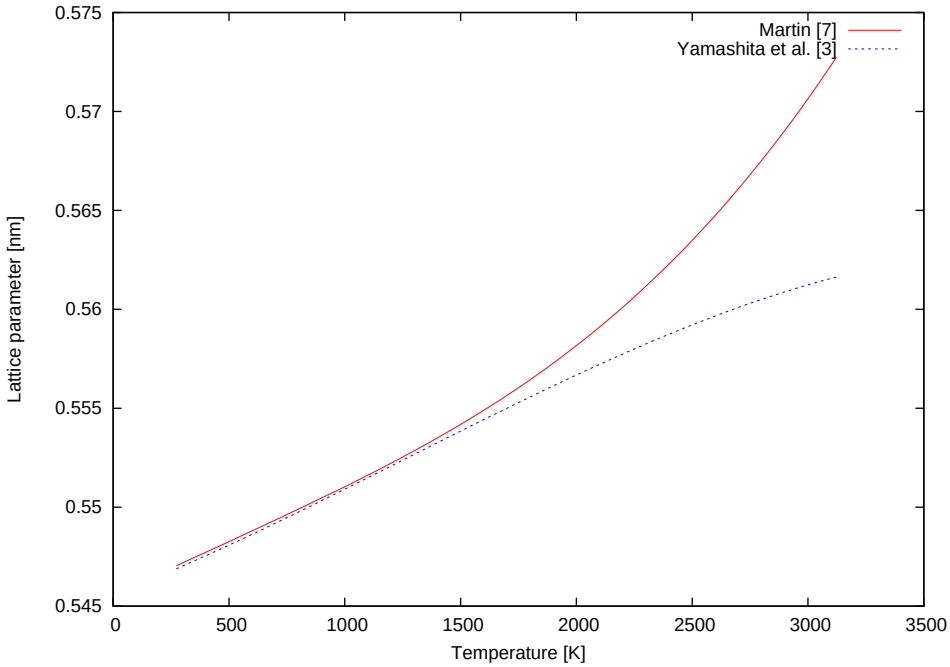

Figure 1. Comparison of different thermal expansion laws for UO_2.

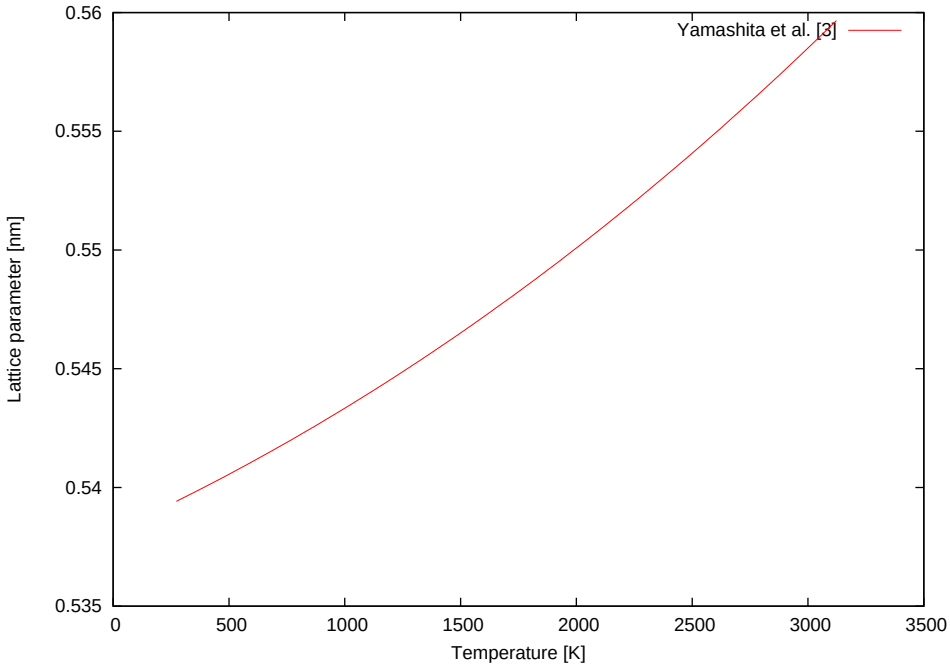

Figure 2. Thermal expansion of PuO_2.