THORIA EHANCEMENT OF NUCLEAR REACTOR SAFETY

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ABSTRACT

The recent nuclear accident in Fukushima demonstrates that a nuclear mishap should be viewed using interdisciplinary tools. For example, the urania fuel melts not only because of enhanced neutron flux but also because its thermal conductivity degrades when it oxidizes. Here we present the application of first principles calculations to evaluate the structural, mechanical and thermal properties of traditional urania fuel and new, inherently safer thoria fuel. Knowledge of the lattice constants for higher oxidation state oxides of uranium is important in nuclear safety analysis, since the formation of U_3O_8 in a defective fuel element can cause cracking or split the fuel sheath after disposal, due to a net 32-36% volume increase. This contrasts with thoria fuel where such oxidation does not occur during accident. The comparison between these two fuels is provided with respect to reactor safety. In an exemplary simulation we show that replacement of urania fuel by thoria prevents central melting in the fuel rod.

Keywords: Thoria, Urania, Nuclear Reactor Safety, Simulations

1. INTRODUCTION

The recent tragic accident in Fukushima clearly illustrates the risks associated with the present design of reactors based on uranium oxide fuel and justifies the research towards a safer fuel. Traditionally, safety protocols focus on reactor physics to evaluate the behavior of a nuclear reactor. However real nuclear accidents (like the recent reactor failure in Fukushima) demonstrate that a nuclear mishap should be viewed using interdisciplinary tools.

For example the fuel melts not only because of enhanced neutron flux but also because its thermal conductivity degrades when fuel oxidation takes place (Lewis et al., 2002). Another study (Szpunar et al., 2001) shows that urania fuel does not start to oxidize until the protective Zircaloy cladding is fully oxidized. However during the oxidation of Zircalloy cladding hydrogen is produced ($Zr+2HO_2 \rightarrow ZrO_2+2H_2$) and it may explode as happened during the Fukushima accident.

Fresh urania fuel has a cubic crystal structure (Fm$\bar{3}$m symmetry observed experimentally (Wyckoff, 1963).

UO_2 has a complex magnetic structure (noncollinear triple-k antiferromagnetic ordering (Wilkins et al., 2006)), which is stable only up to $T_N = 30.8K$ (Frazer et al., 1965), whereas the experimentally available data on structural and mechanical properties are measured in the paramagnetic state. In the paramagnetic state there is no long-range order, but, because 5f electrons are strongly localized, the local moments (1.74 μ_β (Faber et al., 1976; Lander, 1980)) should not be affected much by magnetic ordering.

The most stable uranium oxide is U_3O_8.with the orthorhombic pseudo-hexagonal phase (C2mm) at room temperature and hexagonal phase ($P\bar{6}2m$) that is stable above 400°C (Loopstra, 1970). Interestingly when urania oxidizes initially to U_4O_9 a small shrinking of volume occurs as discussed in (Szpunar and Szpunar, 2004) and is attributed to the reduction of the magnetic moment on uranium. However U_3O_8 has higher volume per U atom than UO_2 and the respective relative linear expansion of urania $\upsilon = 1.36$ (McEachern and Taylor, 1998) and $\upsilon = 1.32$ (Iglesias et al., 1989) was observed experimentally during oxidation to U_3O_8. The created stress (Szpunar et al.,

2013, Szpunar and Szpunar, 2013b) during formation of U_3O_8 affects not only the safety of the nuclear reactor but also the fuel storage facilities as cracking and fragmentation may occur. The oxidation and reduction cycle (OREOX) can also break the fuel sheath (Sullivan and Cox, 1995) due to increasing stress caused by the expansion of oxidized fuel (U_3O_8). This process is also used to produce the powdered uranium oxide for fuel pellet production.

Thoria fuel presents an attractive alternative to traditional urania fuel. It is widely discussed that thoria is not only four times more abundant but also that nuclear reactors based on thorium would be safe, with the risk of reactor core melt-down eliminated due not only to higher thermal conductivity but also a much higher fuel melting temperature: 3651±17 K (Ronchi and Hiernaut, 1996) versus 3120±30 K (Adamson *et al*., 1985) for urania. However it is seldom discussed that thoria does not oxidize to the higher oxidation states like urania. Therefore it has the further benefit that cracking and fragmentation would not occur as happens with urania. Additionally its thermal conductivity does not deteriorate due to oxidation as with urania, (Lewis *et al*., 2002) and these characteristics will be explored here.

1.1. Application of First Principles Calculations

1.1.1. Uranium Oxides

The commonly used Density Functional Theory (DFT) (Kohn and Sham, 1965) within the Local-Density Approximation (LDA) (Ceperley and Alder, 1980) predicts that urania is metallic, while experiment has observed it to be an insulator with a 2.1 eV band gap (Baer and Schoenes, 1980). To address shortcomings of DFT that underestimate the influence of the strong on-site Coulomb repulsion between the strongly correlated 5f electrons, in our work the Hubbard U correction (DFT+U scheme (Cococcioni and Gironcoli, 2005)) is used. CASTEP (Segall *et al*., 2002) calculations with LDA and LDA+U for the effective U value of 2 eV and 3.5 eV were presented (Szpunar, 2012a). The latter value reproduces the best experimental optical band gap of 2.1 eV (Baer and Schoenes, 1980). The shift in energy and the width of the created gap are proportional to the U value, as expected. We observed exactly the same energy shifts and band gaps for two uranium atoms in antiferromagnetic urania with antiparallel magnetic moment orientation (Szpunar, 2013).

1.2. U_3O_8

There are about 67% more oxygen atoms per uranium atom in U_3O_8 than in UO_2. In the P 2m structure all uranium sites are equivalent; therefore when assuming the same symmetry for the electronic structure the simple valency consideration leads to 5.33 noninteger valency of ions (1.33 valence increase from quadrivalent ionic state in UO_2). Therefore as discussed previously (Szpunar *et al*., 2013; Szpunar and Szpunar, 2013b) there is no band gap formed for the U_3O_8 compound unless equivalency symmetry restrictions are removed and it is allowed for uranium ions to have different valency (e.g., from simple model: Two hexavalent and one tetravalent state or two hexavalent and one pentavalent state as indicated in (Yun *et al*., 2011)). Therefore the band gap value: 2.17 eV comparable to the observed value in urania (Baer and Schoenes, 1980) has been found (Szpunar *et al*., 2013; Szpunar and Szpunar, 2013b) in orthorhombic (C2mm) U_3O_8 for U value equal to 6 eV (note: Proposed before in (Geng *et al*., 2007) for urania). However this band gap value depends on the value of Hubbard U. There are no experimental data available about this band gap width.

The most important prediction of the first principles simulation is that there is an increase in the volume of between 35-39% per uranium atom (independent on the used functional) during transformation from UO_2 to U_3O_8 (Szpunar *et al*., 2013; Szpunar and Szpunar, 2013b) and therefore cracking and fragmentation of urania fuel occurs during oxidation (Szpunar, 2013) as also discussed above.

1.3. Thoria Versus Urania Fuel

The crystal structure of thoria is cubic Fm$\bar{3}$m, like urania, but it is nonmagnetic. In contrast to urania, thoria is predicted to be an insulator within the LDA (or GGA) scheme, although the band gap's values are underestimated. However as discussed previously (Szpunar *et al*., 2013) for thoria to represent correctly the wide band gap of 6 eV (Sviridova and Suikovskaya, 1967), the 5f electrons are only pushed up sufficiently when the B3LYP functional (Becke, 1993) is used. This leads to a larger band gap (6.9 eV), in better agreement with experiment (6 eV) than value (4.7 eV) calculated using the LDA+U (6 eV) scheme (Szpunar *et al*., 2013). In our recent research we have studied the mechanical, structural, electronic and optical properties of thoria (Szpunar *et al*., 2013, Szpunar and Szpunar, 2013b). In the following two sections we summarize our preliminary studies using DFT to compare thermal expansion, melting temperatures and minimal thermal conductivity of thoria and urania as presented in more details (Szpunar, 2013).

1.4. Melting Temperature

Recent research (Sanati *et al*., 2011) demonstrated that empirical correlation (Fine *et al*., 1984) between C_{11} elastic constant and melting temperature (T_m) furnishes the latest in good agreement with experiment. **Table 1** shows the evaluated melting temperature for urania and thoria. We used the previously calculated (by us) C_{11} using various functionals (Szpunar *et al*., 2013) and the empirical correlation (Fine *et al*., 1984) Equation (1):

$$T_m[K] = 553 + 5.91 C_{11}[GPa] \qquad (1)$$

We note that this relationship was tested for various cubic metals and compounds and agreement was within 300 K.

The experimental value of the melting temperature of urania (3120±30 K (Adamson *et al*., 1985)) is lower than that evaluated as the most accurate (Bakker *et al*., 1997) value at exact stoichiometry (3651±17 K (Ronchi and Hiernaut, 1996)) and other evaluated melting temperatures between 3323 and 3663 K (Bakker *et al*., 1997). However as **Table 1** shows the estimated melting temperature of thoria is lower than urania and it is underestimated by more than 300 K while for urania agreement is good for most functionals (except of PBE).

1.5. Minimal Thermal Conductivity

The phonon contribution to thermal conductivity decreases with temperature (e.g., for urania Ref. (Lewis *et al*., 2002)) and it is of interest for reactor safety analysis to estimate the minimum thermal conductivity that according to (Cahill *et al*., 1992) is proportional to sound velocity v Equation (2):

$$v = \sqrt{B/\rho} \qquad (2)$$

where, B is bulk modulus and ρ is density. Since thoria and urania have the same structure, the ratio of the minimal thermal conductivity is equal to the ratio of sound velocities and it can be expressed in the previously evaluated (e.g., in (Szpunar *et al*., 2013; Szpunar and Szpunar, 2013a)) bulk moduli (B), lattice constants (a) as Equation (3):

$$\frac{\kappa_{min}^{ThO_2}}{\kappa_{min}^{UO_2}} = \frac{v_s^{ThO_2}}{v_s^{UO_2}} = \sqrt{\frac{B_{ThO_2}(A_U + 2A_O)a_{ThO_2}^3}{B_{UO_2}(A_{Th} + 2A_O)a_{UO_2}^3}}$$
$$= \sqrt{\frac{270 B_{ThO_2} a_{ThO_2}^3}{264 B_{UO_2} a_{UO_2}^3}} \qquad (3)$$

where, $A_{element}$ are atomic masses of the indicated atoms. The calculated ratio of the thermal conductivity of ThO_2 versus UO_2 (phonon contribution) at 1800 K as used in (Bakker *et al*., 1997; Lewis *et al*., 2002) is 1.02 and compares well with the estimated values in **Table 2** for various functionals (LDA (Ceperley and Alder, 1980), WC (Wu and Cohen, 2006), LDA+U (Cococcioni and Gironcoli, 2005) (urania), B3LYP (Becke, 1993) (thoria) and experimental data).

1.6. Thermal Conductivity of Urania and Thoria

1.6.1. Urania

In order to determine the temperature distribution in uranium oxides, the thermal conductivity of the fuel must be known. There are three possible mechanisms of the heat conduction in urania: Lattice vibration (phonons), electron-hole movement (polarons), radiative contributions (photons). The phonon contribution to the thermal conductivity of urania k_{ph} can be represented by the expression (Lucuta *et al*., 1996; Ellis *et al*., 2000):

$$k_{ph} = \frac{1}{A(x) + B(x)T} \quad W\ m^{-1}\ K^{-1} \qquad (4a)$$

where according to Lucuta *et al*. (1996):

$$A(x) = 0.0257 + 3.336x$$
$$B(x) = 0.0002206 - 0.000685x \qquad (4b)$$

Or as proposed by Ellis *et al*. (2000):

$$A(x) = 0.014 - 0.010763\sqrt{x} - 2.3814x$$
$$+ 12.81986\left(\sqrt{x}\right)^3 - 5x^3$$
$$B(x) = 0.0002218 + 0.0002562\sqrt{x} - 0.00064x$$
$$- 0.0036764\left(\sqrt{x}\right)^3 + 0.0173x^3 \qquad (4c)$$

The parameter A in Equation 4c has been slightly modified from the original analysis in order to predict the central melting that was observed in the lower part of the fuel rod in experiment FFO-103 at a linear power rating of 52 kW/m (Lewis *et al*., 2002).

At high temperatures the phonons' contribution to thermal conductivity becomes less important while the reverse is true for the radiative contribution. The radiative term in the current analysis is based on the previously proposed contribution (Hyland, 1983; Reid *et al*., 1997):

$$k_{rad} = 7.59375 \times 10^{-7} (\alpha_R (T))^{-1} T^3 \qquad (5a)$$

Table 1. C11 elastic constants and melting temperature evaluated using Equation 1

Compound	Functionals	C_{11} [GPa]	T_m [K]
ThO$_2$	LDA (Ceperley and Alder, 1980)	385.0	2828.350
	PBE (Perdew et al., 1966)	351.9	2632.729
	PBEsol (Perdew et al., 2008)	370.9	2745.019
	WC (Wu and Cohen, 2006)	370.6	2743.246
	B3LYP (Becke, 1993)	373.1	2758.021
	LDA+U	337.0	2544.670
	Exp.	377.0 (Clausen et al., 1987)	2781.070
	Exp.	367.0 (Macedo et al., 1964)	2721.970
UO$_2$ (AF)	LDA+U	389.2	2853.172
UO$_2$	LDA+U	370.5	2742.655
	LDA (Ceperley and Alder, 1980)	411.9	2987.329
	PBE (Perdew et al., 1966)	318.2	2433.562
	WC	398.0	2905.180
	Exp.	389.0 (Fritz, 1976)	2851.990

Table 2. The ratio of the minimum of the thermal conductivity of thoria versus urania as evaluated using Equation 3

Method	B_{ThO2} [GPa]	a_{ThO2} [nm]	B_{UO2} [GPa]	a_{UO2} [nm]	K_{min} (ThO$_2$/UO$_2$)
LDA	213.4	0.553	229.1	0.53300	1.03
GGA/WC	203.0	0.556	221.7	0.53500	1.02
LDA+U/B3LYP	196.2	0.559	212.5	0.54600	1.01
Experiment	208 (av.)	0.560	208.9	0.54582	1.05
	(Clausen et al., 1987; Macedo et al., 1964)	(Mathews et al., 2000)	(Fritz, 1976)	(Wyckoff, 1963)	

Where:

$$\alpha_R(T) = C_1 \exp(C_2 \times T) \tag{5b}$$

with C_1 equal to 8750 m^{-1} and C_2 having a value of 7.5971×10^{-4}K^{-1}.

Additionally, at high temperature for urania fuel there is a significant electron-hole: (Polaron) transport contribution to the thermal conductivity (Reid et al., 1997; Lewis et al., 2002):

$$k_e = \left(\frac{k_B}{e}\right)^2 T \frac{\sigma_e \sigma_h}{\sigma_e + \sigma_h} \left(\frac{\Delta U}{k_B T}\right)^2 \tag{6a}$$

where, k_B is the Boltzman constant, e is the electron charge, σ_e is partial dc electric conductivity due to electron transport, σ_h is the partial dc electric conductivity due to the hole transport and ΔU is the Mott-Hubbard energy gap. According to (Reid et al., 1997) the above expression for a given temperature and stoichiometric deviation (x) leads to the formula:

$$k_e = C_\sigma \left(\frac{\Delta U}{k_B T}\right)^2 \frac{n p (1 - n - p)}{n + p} e^{-\Delta E/(k_B T)} \tag{6b}$$

where, C_σ is a constant equal to 3.71Wm^{-1}K^{-1}, ΔU is the Mott-Hubbard energy gap equal to 4.33×10^{-19}J, k_B is the Boltzman constant, ΔE is the electron mobility activation energy equal to 4.81×10^{-20}J. The variables p and n are calculated from the conditions of electroneutrality and thermodynamic equilibrium. The molar electron (n) and hole (p) concentration for a given stoichiometric deviation (x) are represented by equations:

$$p = x + \frac{-2\gamma + \sqrt{x^2(1 - 4\gamma) + \gamma}}{1 - 4\gamma} \tag{6c}$$

$$n = p - 2x \tag{6d}$$

Where:

$$\gamma = \exp(-\Delta F / (k_B \times T)) \tag{6e}$$

$$\Delta F = \Delta U - T\Delta S \text{ and } \Delta S = 2.62 \times 10^{-23} J / K \tag{6f}$$

Recently Fink (2000) made recommendations for the thermal conductivity correlation of stoichiometric UO$_2$ and in the recent review (Carbajo et al., 2001) correlation was provided for oxidized urania. In these proposed correlations the high temperature contribution

is fitted by a single (ambipolar) term without separation of the radiative contribution. In contrast to the electronic contribution's dependence on a stoichiometry deviation, as presented here, the recommended (Carbajo *et al*., 2001) high temperature term is not dependent on stoichiometry and therefore the reduction of the thermal conductivity with increasing stoichiometry deviation, as predicted here, may be too conservative. However as shown below, the model presented here for urania can easily be adapted for thoria.

1.7. Thoria

The phonon contribution to the thermal conductivity is expressed by the same equation as for urania (Equation 4a) except that there is no dependence on stoichiometry deviation (x) for parameters A and B and the following values were proposed by Bakker *et al*. (1997): $A = 0.0042$ mKW^{-1} and $B = 0.000225$ mW^{-1}.

Belle and Berman (1984) it was pointed out that there are many uncertainties with evaluation of a radiative contribution to a thermal conductivity as it may depend on the relative mean free path and dimensions of the radiative objects' surface roughness that are not included in the original black body radiation model. The fuel is also not in equilibrium with the adjacent medium into which it is radiating energy. In the present evaluation we assume that the electromagnetic radiation in the thoria and urania fuels contains the same number of quantised electromagnetic waves at the same temperature and the only difference in their radiative conduction originates from the different critical angles of the total internal reflection on the surface of the fuel rods. Both urania and thoria have a higher refraction index (Szpunar and Szpunar, 2013b) than surrounding media, which has an index of refraction equal ~1. Siegel and Howell (2002) the effect of the total internal reflection of radiation on the surface of the body with a refraction index (n) larger than one and adjacent medium with an index of refraction equal to one has been evaluated. A reduction in the radiative heat conduction by n^{-2} was found. Using our previous first principles calculation of the index of refraction (Szpunar *et al*., 2013) the average ratio (wavelength between 200 and 7000 nm) of the calculated index of refraction of thoria and urania was estimated as S_{av} equal to ~0.6877. Therefore we propose here to use Equation (5a-b) with the modification originating from the reduction of the total internal reflection due to lower index of refraction: n ($n_{thoria} = S_{av}n_{uarania}$) of thoria by corresponding factor $(S_{av})^{-2} = 2.11447$ Equation (7):

$$k_{rad} = 16.05677 \times 10^{-7} (\alpha_R (T))^{-1} T^3 \qquad (7)$$

where, the expression (Equation 5b) for $\alpha_R(T)$ is not changed. Since thoria has a reduced index of refraction, the respective radiative contribution is about twice that for urania in fuel rods.

In contrast it was evaluated that the electronic contribution is much smaller for thoria fuel. In the evaluation of the electronic contribution for thoria we set in Equation 6a-f, stoichiometric deviation (x) equal to 0 and increased ΔE, the electron mobility activation energy by a factor of three ($\Delta E_{thoria} = 3\Delta E_{urania}$) as estimated from the ratio of the band gaps (~2 eV for urania and ~6 eV for thoria (Szpunar *et al*., 2013; Szpunar and Szpunar, 2013b)). Since there is exponential dependence of the electronic contribution on the ΔE (Equation 6b) the increase of the activation energy by a factor of three reduces drastically the electronic term.

1.8. Thermal Conductivity of Thoria Versus Urania

In **Fig. 1** we compare the total thermal conductivity of thoria versus urania calculated as a sum of all contributions (phonons, electrons, radiation) Equation (8):

$$k = k_{ph} + k_e + k_{rad} \qquad (8)$$

We will be using here two previously proposed correlations for phonon contribution (Ellis *et al*., 2000; Lucuta *et al*., 1996) for a fully dense urania fuel (Equation 4a-c) with the effect of stoichiometry deviation (x) included and with the electron and radiative contribution terms described by Equation 5-6. They are indicated in **Fig. 1** by dot dot dash and broken line respectively and agree with each other well for stoichiometric fuel. Therefore we only plotted the phonon contribution for stoichiometric urania (triangles) and as indicated it is the only mechanism responsible for thermal conductivity at low temperature. Fink's (2000) correlation (indicated by solid line) was developed for 95% dense urania fuel without the dependence on (x) and it is more conservative at lower temperature while slightly higher thermal conductivity is predicted at high temperatures.

The red dot dash curve represents the total conductivity of thoria, which is calculated as a sum of phonon contribution (Bakker *et al*., 1997), indicated by red triangles and polaron and radiative contribution as described above. It can readily be verified that the thermal conductivity of thoria is much higher at low temperatures than for urania and at high temperatures the phonon contribution decreases significantly, being only a few percent higher as evaluated in **Table 2** for minimal conductivity ratios of thoria versus urania.
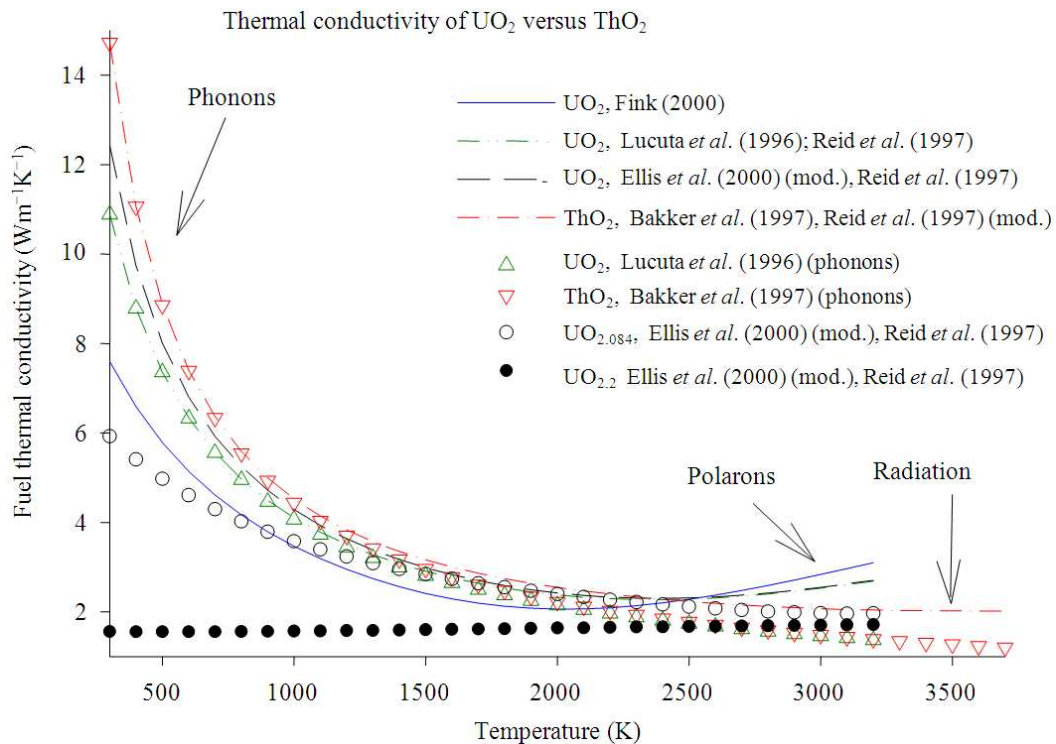
Fig. 1. The calculated thermal conductivity of urania and thoria as a function of temperature. The lines represent the total thermal conductivity (Equation 6) while triangles show phonon contribution (Equation 3a). The primary contribution at high temperature (electronic/polarons for urania and radiation for thoria). Various correlations as described in the text and indicated are used for the phonon contribution while other contributions are calculated using Eqs. 4-a except for the correlation recommended by Fink (2000) indicated by solid red curve. The circles show the reduced thermal con-ductivity of urania for stoichiometry deviation equal to 0.084 (open circles) and 0.2 (solid circles)

In **Fig. 1** it is also illustrated that the thermal conductivity is significantly reduced for urania when oxidation (x = 0.084: Open circles and x = 0.2 solid circles) takes place as also discussed before (Lewis *et al.*, 2002).

In the presented model the thermal conductivity of urania at low stoichiometry deviation and high temperature originates primarily from electronic contribution as indicated in **Fig. 1**. In contrast for thoria, which has a radiative contribution about twice higher, the electronic contribution is negligible and therefore **Fig. 1** indicates that the value of thoria conductivity at high temperature of around 2 $Wm^{-1}K^{-1}$ is radiative. This is in agreement with the value shown in figure 6.24 in (Belle and Berman, 1984) of the flat thermal conductivity of thoria at high temperature around 2 $Wm^{-1}K^{-1}$. The value of 2.33 $Wm^{-1}K^{-1}$ at 2300 K as shown in **Fig. 1** is also in agreement with the 2.5 $Wm^{-1}K^{-1}$ value reported by Hyland (1983) where it is noted also that this thermal conductivity value is maintained up to melting temperature.

1.9. The Temperature Profile in the Defective Fuel Rods

Once the thermal conductivity is known one can calculate the temperature distribution in a cylindrical rod of urania (Lewis *et al.*, 2002) or thoria fuel for a specified volumetric heat generation rate from fission (H) Equation (9):

$$H = \frac{P}{\pi a^2}\left[\frac{(aL^{-1})}{2I_1(aL^{-1})}\right]I_0(rL^{-1}) \tag{9}$$

where, P is linear power (P = 52 kWm^{-1}) and "a" is the radius of the fuel rod (a = 0.006 m). The thermal neutron diffusion Length (L) equal to 1.1 cm for natural urania with the burnup of 8000 MWd/t as estimated in our previous work (Lewis *et al.*, 2002) is also used here. The neutron diffusion length for pure thoria is longer but it will be shorter when doped with Pu or U, therefore for simplicity we assume the same profile for the neutron

flux depression (Equation 9) where I_0 and I_1 are the modified Bessel functions of the first kind.

The temperature profile in the nuclear fuel rod is calculated by solving the steady state heat conduction equation for cylindrical symmetry (Olander, 1976):

$$\frac{1}{r}\frac{d}{dr}\left(rk\frac{dT}{dr}\right)+H=0 \tag{10a}$$

Equation (10a) is subject to the boundary conditions Equation (10b-10c):

$$\frac{dT}{dr}=0, \quad r=0 \tag{10b}$$

$$T=T_s, \quad r=a \tag{10c}$$

where, T_s is the fuel surface temperature, assumed here to be 870 K (Lewis *et al.*, 2002). Here k is the thermal conductivity, which is a function of temperature and consequently depends on the radial position r. Additionally for operating defective nuclear fuel rods that are made from urania, fuel-oxidation may take place and therefore the thermal conductivity will be reduced with increasing stoichiometry deviation (x) as discussed in the previous section and presented in **Fig. 1**. We

assume here the time of origin of simulation to be the time after the zircalloy cladding has fully oxidised since, as calculated before (Szpunar *et al.*, 2001), there is no significant urania fuel oxidation earlier due to a low value of oxygen potential in the presence of hydrogen originating from the oxidation of zirconium in steam. Recently fuel oxidation models in steam atmosphere were evaluated (Szpunar, 2012b) and while a more accurate equation for a steam dissociation was provided it was also demonstrated that there was not much effect on the calculated stoichiometry deviation; therefore we use here the simplified model used previously (Lewis *et al.*, 2002; Szpunar *et al.*, 2001). The estimate of equilibrium stoichiometry deviation (Blackburn, 1973) used here is conservative at high temperatures and the difference in the calculated equilibrium stoichiometry deviation at low temperatures from more conservative models is not significant (Szpunar and Szpunar, 2012).

In **Fig. 2** the temperature profile in the fuel rod is shown as simulated after one day's irradiation time at a linear power of 52 kWm-1. When no oxidation takes place the predicted temperature profiles for urania and thoria are very similar (for the correlations used for thermal conductivity as indicated) and are presented by broken and solid lines for urania and a dot dash line for thoria in **Fig. 2**.
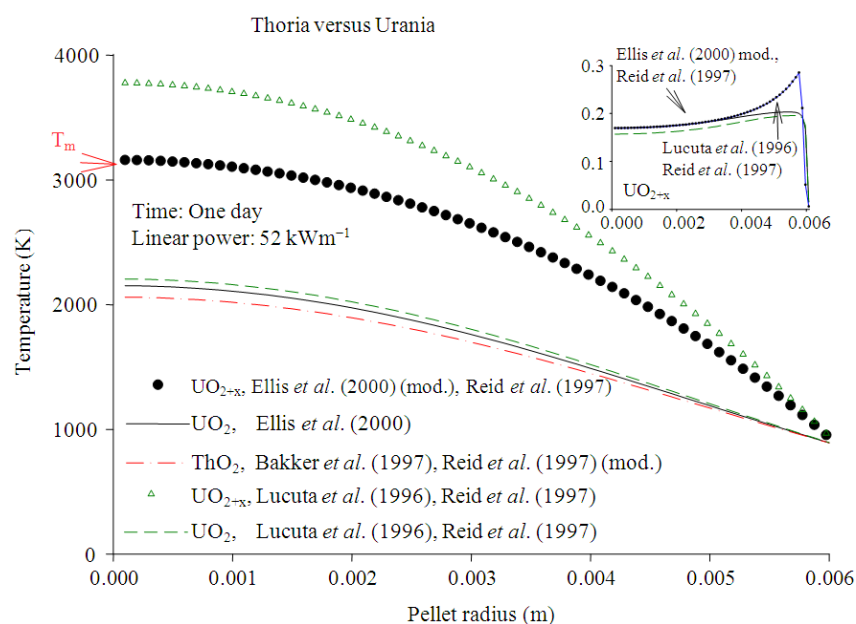

Fig, 2. Predicted fuel temperature profile for stoichiometric thoria and urania (indicated by line). The calculations in a defective fuel rod operating at a linear power of 52 kW/m in which pure steam is present in the gap atmosphere are also shown and indicated by circles and triangles for two different correlations for thermal conductivity as indicated and described in the text. The arrow indicates the melting temperature (Tm) for urania: 3120K (Adamson *et al.*, 1985). The temperature is drastically enhanced when urania fuel oxidation takes place and in the insert the respective stoichiometry deviations are shown as indicated. The pick close to the surface of the pellet is also shown for the results where the oxygen diffusion in the pellet was neglected (solid/dotted line in the insert)
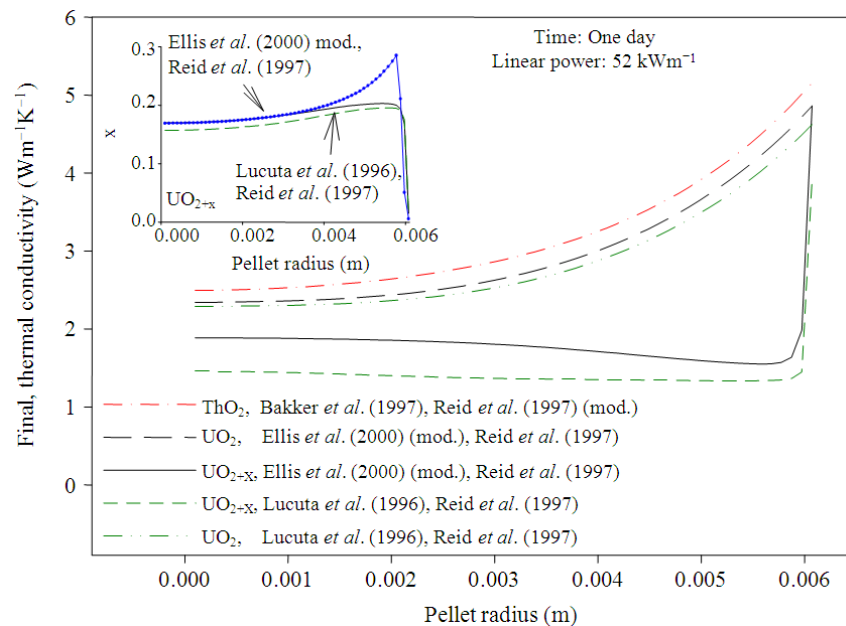
Fig. 3. The calculated thermal conductivity profile in urania and thoria pellet (as a function of the distance from the centre: r) after one day's irradiation at 52 kWm-1 linear power. The lines represent the total thermal conductivity using various correlations as described in the text and indicated. In the insert the respective stoichiometry deviations are shown as indicated. Additionally, indicated by a solid/dotted line with the pick close to the surface, the results for stoichiometry deviation are shown where oxygen diffusion in the pellet was neglected

However when urania fuel oxidation takes place the temperature is drastically increased (indicated by triangles and circles) and central melting in the fuel rod occurs. This can be compared with the melting temperature of urania (T_m), indicated by the arrow, as observed experimentally (Lewis *et al.*, 2002). This temperature increase is due to greatly reduced thermal conductivity as shown in **Fig. 3**. We also note that the correlation proposed by Carbajo *et al.* (2001) predicts an increase of temperature at the centre below melting 60 K. In the inserts of **Fig. 2 and 3** the calculated flat profile of the stoichiometry deviation of urania is also shown after one day. Additionally the dotted line shows that there would be a higher stoichiometry deviation close to the surface of the fuel if the diffusion of oxygen were not included as observed previously (Lewis *et al.*, 2002).

2. CONCLUSION

It is important to include multidisciplinary tools in reactor safety analysis. First principles simulations are useful in assessing the behaviour of nuclear materials especially where there are no experimental data available. They are also useful as a complimentary tool

in reactor safety analysis. Our analysis indicates that the main advantage of using thoria versus urania fuel is that it does not form oxides during accident and therefore there is no undesirable degradation of its thermal conductivity. The fuel melting observed in urania fuel rods during accident with steam ingress is not observed in thoria as its thermal conductivity remains high enough to dissipate excessive heat in the centre.

3. ACKNOWLEDGEMENT

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