Thermal conductivity coefficient UO_2 of theoretical density and regular stoichiometry

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Abstract. The main methodological research results of the thermal conductivity of uranium dioxide (UO_2) were considered. UO_2 is the main fuel of modern nuclear energetic reactors. The assessment of efficiency of the proposed approximations compared to Fink-Ronchi formula at the theoretical values of UO_2 density is proposed. According to this methodology we conducted the analysis of the experimental dependences of the thermal conductivity obtained by domestic authors.

1 Introduction

Nowadays uranium dioxide is the main fuel of nuclear energetic reactors owing to its chemical stability and radioactive resistance. The first accessible reference data on thermal and physical characteristics UO_2 were presented in research work [1] at the end of 60s.

Operating experience and further researches of oxide fuel in 1968-1998 showed significant differences in dependence of thermal conductivity coefficient on temperature $\lambda(T)$, both fuel production technology and further influence on its meaning during reactor work of fission products, porosity changes and stoichiometry composition of fuel [2-4].

In our opinion, important methodical specifics of the research stage of thermal and physical properties UO_2 are:

- introduction of idea of theoretical density UO_2;
- detailed understanding of thermal transfer mechanism to thermal conductivity models of crystal sampling $\lambda(T)$ in the form of sum of phonon or lattice conductivity λ_{ph}, photon or radiation λ_r and electronic conductivity, including ambipolar conductivity, λ_e;
- receiving of the first recommended Harding-Martin's formula [5] for thermal conductivity design UO_2 of theoretical density at temperature ranges 773–3120K which included two main components of thermal conductivity mechanism (pic.1);
- use of integral thermal conductivity for temperature changes design in fuel pellet.

On the whole, at this stage of research insufficiency of experimental data on thermal conductivity UO_2 at high temperatures conditioned by numerous amount of influencing factors and difficulty of making experiments was identified.

There are ranges of temperature changes of fuel pellets in cassettes of medium and maximum reactor power VVER-1000 [6] shown on the graph of thermal conductivity

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coefficient dependence UO_2 on temperature (pic.1) which underline the role of each component of thermal transfer mechanism.

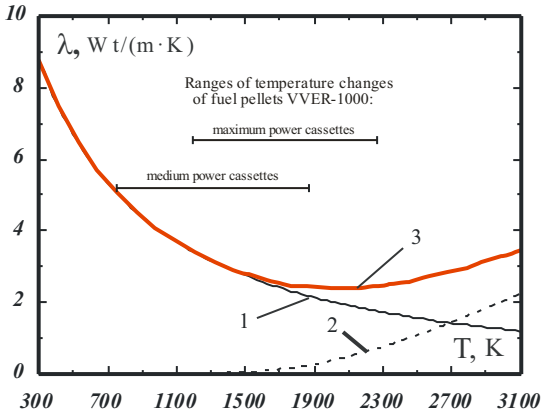

Fig. 1. Thermal conductivity UO_2: 1 – phonon component of thermal conductivity; 2 – electronic component; 3 – full thermal.

Thereafter in 1999-2008 as a result of generalization of more extensive experimental data Ronchi [7] offered a new dependence for ambipolar component. Later Fink generalized other researcher's results and found modified expression for phonon component. He used Ronchi's expression for ambipolar component and received a design formula of thermal conductivity coefficient UO_2 [8,9] which inaccuracy is +10% till the limit of 2000 K, and +20% if the limit is higher. Coefficient 1.158 was introduced in the Fink-Ronchi's formula below for recomputation of the meaning of thermal conductivity samples UO_2 coefficient from 95 to 100% theoretical density [10]:

$$\lambda = 1.158 \cdot \left(\frac{100}{7.5408 + 17.692 \cdot \tau + 3.6142 \cdot \tau^2} + \frac{6400}{\tau^{5/2}} \exp\left(\frac{-16.35}{\tau} \right) \right), \quad (1)$$

where $\tau = (T/1000)K$.

Introduction of theoretical density UO_2 allows detecting efficiency of offered approximations for thermal conductivity coefficient UO_2 design of theoretical density – λ_0. Let us compare design dependences made by Russian scientists in different years with Fink-Ronchi's formula.

2 Comparison of the approximations on the thermal conductivity of UO_2 at the theoretical values of density

An experimental work [11] proved a theoretical dependence on thermal conductivity coefficient UO_2 design presented in a reference book [12]:

$$\lambda_0 = 1.158 \cdot 10^2 \cdot \left(\frac{1}{3.77 + 0.0258 \cdot T} + 1.1 \cdot 10^{-6} \cdot T + 1.01 \cdot 10^{-13} \cdot T^3 \cdot \exp\left(7.2 \cdot 10^{-4} \cdot T\right) \right). \quad (2)$$

There is also an empiric formula there [12]:

$$\lambda_0 = 1.158 \cdot \left(11.5 - 1.14 \cdot 10^{-2} \cdot T + 4.4 \cdot 10^{-6} T^2 - 5 \cdot 10^{-10} \cdot T^3\right). \quad (3)$$

Influence of inaccuracy from stoichiometry composition UO_{2+x} on thermal conductivity was researched in work [13]. As a consequence, the following dependence led to theoretical density of uranium dioxide was received in the range of temperatures 300-2000K.

$$\lambda_0(x,T) = 1.158\cdot 10^2 \cdot \left(4.5 + 43\cdot x^{1/2} + 0.0219\cdot T - 0.033\cdot x\cdot T\right)^{-1}. \quad (4)$$

Approximation formula [14] given below is shown without specified conditions of application.

$$\lambda = \frac{5500}{560+T} + 0.942\cdot 10^{-10}\cdot T^3. \quad (5)$$

Comparison of formulas (2)–(5) received before 2003 and led to theoretical density UO_2 with Fink-Ronchi's dependence is presented in picture 2; it shows significant differences.

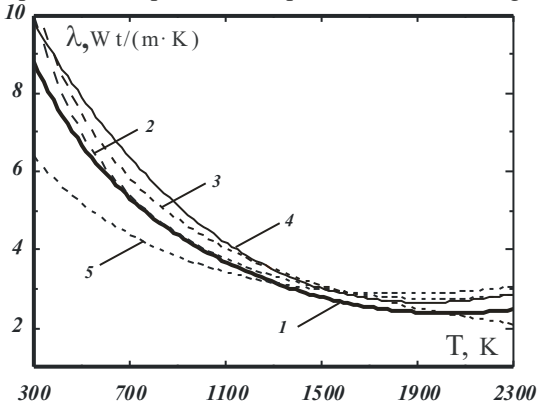

Fig. 2. Comparison of approximations on thermal conductivity UO_2: 1 – Fink-Ronchi's formula (1) [8]; 2 – formula (2), data [11,12]; 3 – formula (3) [13]; 4 – empiric dependence (4) [12]; 5 – formula (5) [14].

In the succeeding reference books and books after 2003 there is move towards standard formula (1). There is empiric dependence in work [15]

$$\lambda_{norm} = \frac{4820}{351+T} + 2.434\cdot 10^{-14}\cdot T^4, \quad (6)$$

in which it is easy to detect a significant difference from Fink-Ronchi's formula led to theoretical density UO_2 (1) (see fig.3, curve 2) while initial conditions of its use were not stipulated. Dependence (1) becomes general in work [16] and further a design formula becomes based on it [17] which includes medium fuel burning-out B, MW·day/kgU:

$$\lambda(B,T) = 1.158\cdot\left\{\left[\begin{array}{l}\left(0.1148 + 3.0864\cdot 10^{-3}\cdot B\right) + \\ 2.475\cdot 10^{-4}\cdot\left(1 - 2.94\cdot 10^{-3}\cdot B\right)\cdot\left(T - 273.15\right)\end{array}\right]^{-1} + \right.$$
$$\left. + 0.0132\cdot\exp\left(1.88\cdot 10^{-3}\cdot\left(T - 273.15\right)\right)\right\}. \quad (7)$$

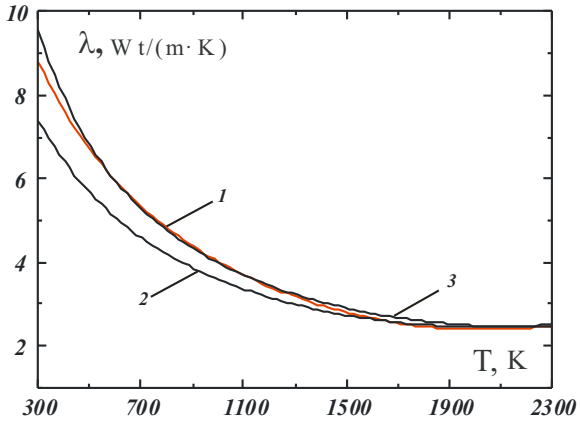

Fig. 3. Comparison of design formulas on thermal conductivity UO_2: 1 – Fink-Ronchi's formula (1) [8,16]; 2 –formula (6), [15]; 3 – formula (7) [17].

Thus, analysis of comparison of experimental dependences of thermal conductivity coefficient UO_2 made by different authors and Fink-Ronchi's formula at theoretical density of fuel allowed:

- to identify significant differences in input of some mechanisms of thermal conductivity assessment with the growth of fuel temperature;
- to detect a base limit of uranium dioxide thermal conductivity change making it possible to clearly distinguish porosity influence, nonstoichiometry and burning-out at present.

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