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Thermo-Physical Properties of Dispersion Nuclear Fuel for a New-Generation Reactors: A Computational Approach

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Abstract. Tomsk Polytechnic University is conducting a series of experiments to develop a nuclear fuel manufacturing technology required for a new-generation innovative nuclear systems. In this study, the thermal conductivity and thermophysical properties were calculated for the proposed nuclear fuel, which are necessary before proceeding to the construction stage. The cylindrical fuel compact consisting of spherical coated constituents (Pu,Th)O₂ of BISO type, were sintered together within a graphite matrix (C). Also, a composite material made of nuclear fuel (Pu,Th)O₂ and refractory oxides BeO/MgO, were prepared using plasma-chemical synthesis method. The development of a fuel pellet with desired physical properties that can be exposed for a long time under irradiation in a high-temperature gas-cooled thorium reactor core is the main technological advantage of the present study.

INTRODUCTION

Since the oxide ceramic nuclear fuels, which are currently used in power reactors, have a number of drawbacks [1], the dispersion nuclear fuels are considered as potential candidates for the new-generation reactors [1-4]. The advantages of the dispersion fuels, in comparison with the traditional oxide fuels, are their high thermal conductivity, radiation and thermal resistance, increased service life and a deeper burn-up of the fissile isotope. The most common type of dispersion nuclear fuel is micro-encapsulated fuel dispersed in a densely sintered graphite matrix (C) or in a matrix consisting of silicon carbide (SiC). This nuclear fuel, which is characterized by the absence of direct contacts between spherical constituents (i.e., coated particles), has improved mechanical, thermal and neutron-physical properties, and the existing production technology allows one to manufacture a fuel compact with a fraction of dispersed coated constituents, \( \phi_f = \frac{N_{\text{particles}} \times V_{\text{particle}}}{V_{\text{matrix}}} \) 37–40 %.

However, the common drawbacks in fabrication technologies of dispersion nuclear fuels are: multi-stage fabrication process, inhomogeneous distribution of the fuel phase, the need for a large number of chemicals and high-energy intensity of production. A new and promising technology for manufacturing dispersion nuclear fuel is plasma-chemical synthesis, which allows the synthesis of fuel compacts with desired physical properties.

Tomsk Polytechnic University is conducting a series of experiments [5] on developing a technology for manufacturing the nuclear fuels using the plasma-chemical methods for nuclear innovation systems of the 4th generation [6-8]. The main advantages of the proposed technology are: single-stage fabrication process, low-energy intensity, and the ability to influence both the size and morphology of the constituents. The composite materials synthesized by the proposed technology has improved thermal conductivity and good neutron-physical properties; Besides, the fuel phases and the matrix are uniformly synthesized in the volume. In this case, (Pu,Th)O₂
and BeO/MgO constituents are synthesized in commonly used spherical and cubic forms, such that their sizes do not exceed 110 nm [5].

The present work aims to develop a practical methodology for studying the thermophysical properties of a fuel with a complex internal structure. Two different conceptual designs for the proposed fuels are introduced: The first fuel type is a compact made of BISO (Bistructural ISOtropic) spherical particles (Pu,Th)O$_2$ sintered with graphite matrix, whilst the second type is a new composite material consisting (Pu,Th)O$_2$ and refractory oxides BeO or MgO.

The thermophysical properties analysis of the proposed fuels is a necessary step before manufacturing a nuclear fuel capable of tolerating a long-time (not less than 7 years) irradiation with epithermal and fast neutron spectra of a high-temperature gas cooled thorium reactor plant (HGTRU) [6,8] without changing its physical properties throughout the whole cycle of irradiation.

**MATERIALS AND METHODS**

**Calculation of Effective Thermal Conductivity of BISO Fuels**

There are many theoretical models for calculating the thermal conductivity of porous materials containing dispersed constituents of different configurations and compositions. In order to find the effective thermal conductivity (ETC) of the BISO fuel compacts, $k_e$, it is generally recommended to use models discussed in [9-19]. A scientific review and a brief description of the models which are used in thermal conductivity calculations have been presented in [18,19]. In order to find the ETC, the following models were used: Maxwell model [9, 10], the Effective Medium Theory (EMT) [11,12], the modified Zehner and Schlünder model [13,14], and the Chiew and Glandt model [15,16].

Maxwell's equation [9] is one of the most well-known equations for finding the thermal conductivity of dispersed media; it provides an exact solution for the thermal conductivity of particles located in a uniform matrix. The theory of effective medium [11] is also a very popular model for describing the thermal conductivity of dispersed media. Both EMT and Maxwell models do not consider the configuration of the dispersed phase, however, they assume that the particles in the matrix volume are randomly distributed. The Zener-Schlünder model can be used for studying the ETC assuming that the particles are in contact with each other. In this study, it has been decided to use a modified Zener-Schlünder equation, which results in a more accurate solution to the heat equation. A modified Zener-Schlünder equation was first obtained by C.T. Hsu in [17]. Although, Chiew & Glandt model can be used to find the thermal conductivity of the particles in a homogeneous medium, like other models, it does not consider the configuration of the dispersed phase.

**Effective Thermal Conductivity of BISO fuel compacts**

Following the recommendations of [18,19], similar numerical calculations were performed to determine the ETC of the fuel compact, where $\omega_f$ (i.e., the volume fraction of fuel compact) varies from 10 to 20%.

The choice of the particle volume fraction $\omega_f$ in a fuel compact was discussed earlier in [20, 21], which is normally based on the neutron-physical studies of the HGTRU core with a fuel compact of type 1017 (Fig. 1a). Fig. 1b illustrates the 3D model of the fuel compact [8,20], the calculated model of the equivalent reactor cell [8], and also the results of neutron-physical studies (Fig. 1c, [20]).
As seen in Fig. 1c [20], similar to the work undertaken in [22], an increase of more than 17–18 % in $\omega_f$ is reasonable only for the hybrid HGTRU core. According to the desired functional dependence, $k_d(\omega_f)$, one may predict the ETC values for the proposed fuel compact, then, the numerical results can be compared with the analytical models discussed above.

**Unit Cell Model**

The numerical calculations of the present stage were performed with the COMSOL Multiphysics program [23]. Three different configurations could be associated with the unit cell model: (1) Simple cubic (SC), (2) Body-centered cubic (BCC), and (3) Face-centered cubic (FCC). The geometrical configurations of all computational models are shown in Fig. 2.

It is worth studying the variation of the ETC of the fuel compact when the fraction of the dispersed phase, $\omega_f$, changes. Since the number of particles in a cell (e.g., in an FCC) does not change, the total particle volume ($V = N_{\text{particles}} \times V_{\text{particle}}$) is equivalent to the volumes of four individual particles, therefore, the variable size will be the cell volume. The cell volume for a cube of the face area, $a$, and the desired value fraction, $\omega_f$, can be related via $a = 2r \times \left(2\pi/3\omega_f\right)^{1/3}$. The simulation was carried out for a configuration consisting of five unit cells, vertically arranged one behind the other (Fig. 3).
The advantage of this configuration is that one can ignore the change in heat flux and temperature in the central cell (i.e., the highlighted cell). In order to calculate the ETC, the values of thermal conductivity of the kernel \( k(\text{Th}_{0.3}\text{Pu}_{0.3}\text{O}_2) = 1.82 \, \text{W/(m} \times \text{K)} \) [24], coatings \( k(\text{PyC}) = 5.7 \, \text{W/(m} \times \text{K)} \) [25,26], \( k(\text{Ti}_2\text{SiC}_3) = 30.98 \, \text{W/(m} \times \text{K)} \) [26]) and matrix \( k(\text{graphite}) = 52.27 \, \text{W/(m} \times \text{K)} \) [27]) were taken at 1670 K temperature.

The ETC \( k_e \) value for the above models was calculated with COMSOL [23,28]. The thermophysical parameters calculations are basically undertaken using the non-stationary heat conduction equation. The initial data for such calculations are normally the thermal conductivity values of the fuel components and also the temperature. The code calculates the value of heat flow, \( Q \), and the surface temperature \( T \), for any configuration. The ETC value of the fuel compact, for example, corresponding to the Unit Cell model (Fig. 3) in one-dimensional stationary case is calculated through the following equation:

\[
k_e = \frac{a q_v + 2Q_{\text{bot}}}{2(T_{\text{bot}} - T_{\text{top}})}
\]

where \( T \) is the temperature, \( t \) is the time, \( z \) is the \( z \)-coordinate of the point, \( k_e \) is ETC and \( q_v \) is the power of internal source. Having incorporated a simple mathematical transformations, the appropriate boundary conditions and the previously calculated \( Q \) and \( T \) values, one may derive the following calculation formula:

\[
k_e = \frac{a Q_{\text{bot}}}{T_{\text{bot}} - T_{\text{top}}},
\]

where \( T_{\text{top}} \) is the top surface temperature of the cell, \( T_{\text{bot}} \) and \( Q_{\text{bot}} \) are the bottom surface temperature and the heat flux of the cell, respectively.

According to [18,19], the heat generation in cores can be ignored. This assumption provides a simpler correlation, which facilitates the \( k_e \) estimation with the necessary practical accuracy. Therefore, by neglecting the heat generation in the kernels \( (q_v = 0) \), the above equation is simplified as:

\[
k_e = a Q_{\text{bot}}(T_{\text{bot}} - T_{\text{top}}),
\]
The calculation formulas for all other models may be derived similarly.

**Random Model**

In the unit cell model, all particles are arranged in a strict order, and the particle centers are located in the cell nodes, such that the particle centers form a regular periodic sequence. Since in reality, the coated particles are randomly oriented in the compact (Fig. 1a), it was decided to use the model proposed in [18], which takes into account the random orientation of particles. Fig. 4 shows the random model configuration with randomly generated particle centers (the front faces are not shown in the figure for convenience). The model is a square base prism (Fig. 4), with square side and height related through \( h = 3 \times a \). There are 100 particles inside the prism, such that 10 particles are on each side surface, and 5 on the lower and upper bases (Fig. 4). Since the particles on the surfaces can share only half of their volumes, the total particle volumes in the prism are equivalent to 125 individual volumes. In this model, the variable is the volume of the prism with side \( a \) and height \( h \). The base side of the prism, \( a \), and its height, \( h \), are related to \( \omega_f \) via \( a = r \times (500 \omega_f / 9)^{1/3} \), \( h = r \times (1500 \omega_f / \omega_f)^{1/3} \).

**Lichtenecker Model**

The plasma-chemical methods are basically utilized to synthesize materials such that the largest particles do not exceed 100 nm. Such a composite material is a nano-powder consist of fuel \((\text{Pu, Th})_2\text{O}_2\) and refractory oxide \((\text{BeO, MgO})\) particles. It is normally a homogenous mixture that can be used to obtain the calculated dependence of \( k_c(\omega_f) \) within the generalized Lichternecker model [29]. This conclusion was previously reported by Novoselov et al. in [5], where the thermal conductivity calculations of metal-oxide compounds were carried out using a number of models discussed in [29].

**RESULTS AND DISCUSSION**

Fig. 5a and 5b show that the ETC of the composite material \(((\text{Pu, Th})_2\text{O}_2 - \text{BeO/MgO})\) is less than that of BISO fuel compact. The ETC of \(((\text{Pu, Th})_2\text{O}_2 - \text{MgO})\) with \( \omega_f = 17 \% \) is more than 2.5 times higher than that of ceramic \((U, \text{Pu})_2\text{O}_2 (k((\text{Th}_{0.5}\text{Pu}_{0.5})_2\text{O}_2) = 1.82 \text{ W/(m × K)} [24] \) (Fig. 5b), and the ETC of \(((\text{Pu, Th})_2\text{O}_2 - \text{BeO})\) is higher than that of ceramic \((U, \text{Pu})_2\text{O}_2\) in more than 10 times. Having taken into consideration that the World annual production of Be is about 150 tons, the metal oxide compound \(((\text{Pu, Th})_2\text{O}_2 - \text{BeO})\), with necessary quantities, is a potential alternative to traditional ceramics, which is used as fuel in power reactors of the LWR type.

It is also worth noting that BISO fuel compacts, as well as composite materials, synthesized by plasma-chemical method; do not exhibit appropriate behavior in the active zone of the HGTRU. High operating temperatures \((1000–1670 \text{ K})\) and very large neutron fluence \( F = (\varphi_{\text{nu}} \times T) = 2.07 \times 10^{26} \text{ n·cm}^{-2}\) with very long irradiation time \((T = 3000 \text{ days})\) substantially reduce the ETC by more than 2 times.

However, the proposed plasma chemical synthesis actively influence the physical properties of the synthesized fuel composition, which hopefully in the near future, facilitate the production of materials with the capability to resist the epithermal and fast neutrons spectrum of the HGTRU at a working fuel temperature of 1000 to 1700 K and a neutron flux of \( \varphi_{\text{nu}} = 8.14 \times 10^{13} \text{ n·cm}^{-2·s} \) (Note should be taken that 90.59\% of neutrons have the energies ranging from 4 eV to 10.5 MeV).
CONCLUSIONS

In the present study, a series of computer calculations has been undertaken to determine the thermal conductivity characteristics for the proposed nuclear fuels of two different conceptual designs. A cylindrical fuel compact consisting of spherical (Pu,Th)O$_2$ particles of BISO type, sintered together with a graphite matrix, and a new composite material synthesized by plasma chemical synthesis were investigated. Studies show that at a fuel temperature of 1700 K, the ETC of BISO fuel compact with $\varphi_f = 17\%$ is 3.8 times higher than that of the metal oxide compound ((Pu,Th)O$_2$-BeO) with the same $\varphi_f$ value. As far as the thermophysical properties are concerned, such a fuel compact is preferable for operation in the neutron spectrum of the HGTRU. However, note should be taken that the final decision on the use of composite material in the reactor can only be made after a series of neutron-physical experiments are performed. The plasma-chemical synthesis methods may produce the particle size not exceeding 100 nm. Such a composite material is a nano-powder and it is practically a homogeneous mixture, which considerably simplifies post-operation neutron-physical and radiation characteristics studies.

According to numerical calculations, the following conclusions may be drawn:

1. Three different cell types discussed in the present study give almost identical results (Fig. 5a), in which the model takes into account the layered structure of the fuel particles, however, it does not consider their random arrangement in the matrix of the fuel compact.

2. In the present study, the Maxwell and the Chiew & Glandt models which are in good agreement with the unit cell model can be used to obtain accurate and reliable results.

3. The random model is the most advanced and accurate model, because it takes into account not only the random particle arrangement in the matrix, but also their configurations. For the fuel compact and the particle type of the present investigation, the random model gives a result that agrees well with the EMT, but it is more expensive (time consuming and complicated) than the calculation methods discussed above.

4. At a fuel temperature of 1700 K, the ETC of BISO fuel compact is 3.8 times higher than that of metal oxide compound ((Pu,Th)O$_2$-BeO) synthesized by the plasma-chemical method, and therefore, the fuel compact is a more preferred choice for the operation at HGTRU. However, the final selection of a particular fuel type for the HGTRU can only be made after a series of neutron-physical experiments is performed on the composite material.

5. The fuel composition synthesized by the plasma-chemical method is a multi-component nano-sized particle mixture, for which the homogeneous models can be incorporated. Both thermal physics and neutron-physical calculations can be undertaken to study the properties of the synthesized compositions in order to propose the materials that can be used in the core of the HGTRU for a long operation time.
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REFERENCES

7. The project for constructing a Small Modular Reactor, [Electron resource], Available at: https://www.inl.gov/article/frequently-asked-questions (accessed 11.08.2018).