COMPARISON BETWEEN EXPERIMENTAL AND STOCHASTIC MODELS FOR EVALUATING THERMAL DIFFUSIVITY OF NUCLEAR FUEL BY THE FLASH METHOD

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ABSTRACT

The thermal diffusivity of uranium dioxide pellets, manufactured without and with addition of beryllium oxide, in order to increase its thermal conductivity, was determined by the flash laser method at room temperature. The thermal diffusivities were estimated using Parker original model and a mathematical stochastic heat diffusion model, based on Inverse Heat Conduction Problems and Monte Carlo Method. The resulting diffusivities were compared. The purpose is to show the accuracy of the mathematical model and to support a research project in progress at CDTN-Centro de Desenvolvimento da Tecnologia Nuclear (Nuclear Technology Development Center), about a new type of fuel manufactured from uranium dioxide kernels with beryllium oxide filling the voids between the kernels. The addition of beryllium oxide is expected to improve the fuel thermal conductivity, in order to avoid a premature fuel pellet degradation and, as a consequence, the fuel remains longer in the reactor causing a large economy. The experiments carried out on the materials demonstrate good agreement between the Parker original model and the mathematical model, and that the thermal conductivity of UO₂ increases when it is added the beryllium oxide. Both the GUM uncertainty framework and a Monte Carlo method were applied.

1. INTRODUCTION

The uranium dioxide is the most used substance as nuclear reactor fuel for presenting many advantages such as: high stability even when it is in contact with water in high temperatures, high fusion point, and high capacity to retain fission products. The conventional fuel is made with ceramic sintered pellets of uranium dioxide stacked inside fuel rods, and presents disadvantages because its low thermal conductivity causes a premature degradation of the fuel due to the large resulting pellet temperature gradient. Besides, the thermal conductivity decreases further as the fuel burns, what limits the fuel operational lifetime. A research project about a new kind of fuel pellets fabricated with uranium dioxide kernels and beryllium oxide filling the empty spaces between them is in progress at CDTN [1]. This fuel has a great advantage because of its higher thermal conductivity in relation to the conventional fuel and as a consequence the fuel remains longer in the reactor causing a large economy. Pellets of this kind had their thermal diffusivity measured by the Parker original...
model [2] and by a stochastic heat diffusion model, based on Inverse Heat Conduction Problems (IHCP) and Monte Carlo Method [3], to compare with the thermal diffusivity of uranium dioxide pellets manufactured with the same kernel batch, but without addition of beryllium oxide, in order to furnish subsides to this CDTN’s research project.

The Parker original model has been considered by several Metrology National Institutes and other organizations as a standard method for the measurement of thermophysical properties of solids. Due its experimental obstacles the original flash method has been reviewed and some alternative and effective approaches are proposed [4,5,6,7] A stochastic modeling has been developed and validated by Standard Samples. Inverse Heat Conduction Problems (IHCPs) solved by Finite Volumes technique were applied to the measurement process with real initial and boundary conditions [3]. Uncertainties of both methods have been estimated according to the ISO/BIPM Guide to the Expression of Uncertainty in Measurement [8] and Monte Carlo Method [9]. The objective of uncertainty evaluation is to quantify the extent and nature of the knowledge of the output quantity given the model of the system. Knowledge of the model input quantities is encoded by the assignment of probability density functions (PDFs) to those quantities. A main requirement is to ascribe to the output quantity a so-called coverage interval that contains a specified proportion, e.g., 95 %, of the distribution of values that could reasonably be attributed to that quantity. There are some limitations and assumptions inherent in the GUM uncertainty framework. In the context of uncertainty evaluation, Monte Carlo is a sampling technique that provides an implementation of the propagation of distributions: the process is undertaken numerically rather than analytically. The technique is also useful for validating the results returned by the application of the GUM uncertainty framework [10].

2. METHODOLOGY

2.1. Uranium Dioxide Kernels and Fuel Pellets Fabrication

The uranium dioxide kernels were produced by the sol-gel process developed by Nukem/Germany that was absorbed, transferred and implemented at the CDTN’s nuclear fuel laboratories [11,12,13]. The process was developed to fabricate fuel elements for high temperature gas cooled reactors. But it was adapted in order to fabricate fuel pellets for pressurized water reactors too [14,15,16,17,18]. By this process a uranium nitrate solution is transformed into spherical droplets that become hard in reaction with ammonium gas and are collected in an ammonium hydroxide solution. In the subsequent steps, the kernels are washed, dried, calcined and sintered. To extract the ammonium nitrate, several washings are made in equipment that revolves the kernels intensely for one hour each washing, up to verify that there is no more ammonium nitrate in the served water, because the presence of ammonium nitrate in the kernels cause its destruction in the subsequent thermal treatment. The drying step was performed at 160 °C for 16 h and the calcination, at 800 °C for 3 h, followed by reduction at 650 °C for 4 h in hydrogen atmosphere, and by passivation under CO₂ atmosphere during the oven cooling down.

Beryllium oxide furnish by Sigma-Aldrich was mixed with the uranium dioxide kernels with a content of 14 and 7 weight percent. To revolve the mixture for 2 h, it was used the same equipment that is used to wash the gel kernels after gelation. The kernels were pressed in pellets using an especial model of hydraulic press developed at CDTN [19,20,21]. The die
used has a diameter of 11.1 mm, the system works with a floating die in order to obtain double effect, and it is used lubricant only on the die surface. The oil used as lubricant was Petrobrás CL-OF-130. The green pellets height and diameter were measured by a micrometer and the mass was obtained with an analytical balance in order to determine geometrically its green density. Fig. 1 shows the uranium dioxide mixed with beryllium oxide pellet microstructure that was obtained by optical microscopy (100x). The light regions in the image correspond to the UO$_2$ phase, the darker ones are the BeO phase.

![Figure 1 – Microstructure of the uranium dioxide and beryllium oxide obtained pellet.](image)

### 2.2. Pellets Density and Open Porosity Measurements

To measure the density and open porosity of the sintered pellets, it was used the xylol penetration-immersion method developed by the Kraftwerk Union research center UO$_2$ laboratory in Erlangen / Germany [22,23,24], which was absorbed, transferred and implemented in the CDTN’s nuclear fuel laboratories [25,26]. The method permits to obtain the open porosity in absolute (% V – percentage of the pellet volume) and relative terms (% P – percentage of the total porosity that is open).

### 2.3. Thermal Diffusivity Measurements

For thermal diffusivity measurement of the pellets were utilized the flash laser method and an in house made bench to apply the flash method according the norm ASTM-E-1461-07 [27,28]. The flash method for measuring thermal diffusivity has been increasingly used since its introduction in 1961, by Parker et al [2]. Fig. 2 schematically presents the experimental apparatus of CDTN for thermophysical properties measurements based on the Flash Method. It consists of CO$_2$ laser working at 10.6 µm wave length. A pulse of energy is applied into the sample and usually set to keep the sample temperature rise below 3 °C. An infrared thermometer measures the transient temperature and the thermal radiance signal is digitized using a 16 bits A/D card. A Lab View programming is used to acquire data. The sample
(8mm in diameter and about 2.5 mm thickness) is placed in a vacuum furnace and isothermally heated. The sample holder consists of three molybdenum screws that fix the sample in vertical position in the central zone of the furnace. The system allows the irradiation of the sample in its frontal face obtaining a register of the temperature transient on the sample opposite face from which the thermal diffusivity $\alpha$ is calculated by the following equation:

$$\alpha = \frac{1.37 \cdot L^2}{\pi^2 \cdot t_{1/2}}$$

where $t_{1/2}$ is the time in which the rear face temperature rise reaches one half of its maximum value and $L$ is the sample thickness.

From the measured data, the thermal conductivity $\lambda$ of the material is determined according to the following equation:

$$\lambda = \alpha \cdot C_p \cdot \rho$$

where $C_p$ is the specific heat and $\rho$ the sample density.

![Laboratory experimental apparatus for thermophysical properties measurements.](image)

2.4. Mathematical model

When the material is uniformly stimulated on its whole front face, heat transfer within the material can be considered one-directional. The model is given by the solution of the one-dimensional heat transfer equation [3]. The experimental procedure considers the mathematical domain represented on Fig. 3. The computer algorithm was developed and implemented using the Compaq Visual FORTRAN platform.
The direct numerical solutions to the thermal diffusion process in the sample are obtained by the Finite Volume Method [29] through CONDUCT.for subroutine (Fig 4).

In the FLASH.for subroutine are implemented the Monte Carlo Method (MCM) and the solutions for Inverse Heat Conduction Problems. The ARRANGE.for subroutine establishes a range of values expected for all inputs parameters of the model based on its initial previous evaluation. The ADAPT.for subroutine implements all initial and boundary conditions. The generation of random draws from Uniform, Gaussian or Triangular distributions is done in the subroutine NORMAL.for. This methodology reduces the degree of model simplification, providing results with improved physical meaning and uncertainty reduction. An optimal inverse solution for the problem is obtained. By using the optimal values evaluated for the input parameters of the model, the thermophysical properties and uncertainties are obtained as results.

Fifteen different sets of temperature rise were generated by a computer in order to demonstrate the mathematical model. In this example, the stability and accuracy of the
All uncertainty sources were quantified and appropriate probability distribution functions (PDFs) assigned to the corresponding input quantities. All PDFs were based on Gaussian or rectangular distributions.

### 3. RESULTS

The table 1 summarized the parameters used to fabricate the pellets and the characterizations results. The uranium dioxide theoretical density (TD) was considered to be 10.96 g∙cm\(^{-3}\). The theoretical density of UO\(_2\)+7% Be0 was considered to be 10.40 g∙cm\(^{-3}\).

<table>
<thead>
<tr>
<th>Material</th>
<th>Compaction Pressure</th>
<th>Sintered Density</th>
<th>Percent of Theoretical Density</th>
<th>Open Porosity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MPa</td>
<td>g cm(^{-3})</td>
<td>%</td>
<td>% V</td>
</tr>
<tr>
<td>UO(_2)</td>
<td>300</td>
<td>9.90</td>
<td>90.3</td>
<td>6.8</td>
</tr>
<tr>
<td>UO(_2)+7% Be0</td>
<td>300</td>
<td>7.97</td>
<td>76.6</td>
<td>16.7</td>
</tr>
</tbody>
</table>

In table 2 and 3 are presented the results of thermal diffusivity and thermal conductivity obtained by original Parker’s model and mathematical model, respectively.

### Table 2. Thermal diffusivity of dioxide samples, without and with addition of beryllium oxide at 20 °C.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Thermal Diffusivity / mm(^2)s(^{-1})</th>
<th>Original Parker’s model</th>
<th>Mathematical Model</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean value</td>
<td>Standard uncertainty</td>
<td>Endpoints of 95% coverage interval</td>
</tr>
<tr>
<td>UO(_2)</td>
<td>2.44</td>
<td>0.11</td>
<td>0.22</td>
</tr>
<tr>
<td>UO(_2)+7% Be0</td>
<td>2.58</td>
<td>0.12</td>
<td>0.23</td>
</tr>
</tbody>
</table>
Table 3. Thermal conductivity of dioxide samples, without and with addition of beryllium oxide at 20 °C.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Original Parker’s model</th>
<th>Mathemathical Model</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean value</td>
<td>Standard uncertainty</td>
</tr>
<tr>
<td>UO₂</td>
<td>6.24</td>
<td>0.31</td>
</tr>
<tr>
<td>UO₂+7% Be0</td>
<td>7.08</td>
<td>0.36</td>
</tr>
</tbody>
</table>

The results indicate the increase when it is added the beryllium oxide at both quantities. The thermal conductivity of the pellet UO₂ is lower than the reported values in literature [30,31,32,33], because its density is not 95 % theoretical density, but only 85.1 % theoretical density that is, its higher porosity results in a lower thermal conductivity. It is planned to make pellets with higher density (95 % theoretical density), and with lower content of beryllium oxide, and to determine by x-ray, the resulted composition of these pellets.

The results show good agreement between the values obtained by original Parker’s model and by mathematical model. The standards uncertainties of the thermal diffusivity in both samples provided by the law of propagation of uncertainty are 4.5 % and 4.7%. Those provided by the described Mathematical model obtained by Monte Carlo method are 3.4 %, in both samples. The value of UO₂ sample thermal conductivity (6.43 W·m⁻¹·K⁻¹) obtained by Monte Carlo method is 3 % larger than that (6.43 W·m⁻¹·K⁻¹) obtained by the original Parker’s model. The standards uncertainties of the thermal conductivity in both samples provided by the law of propagation of uncertainty are 5.0 % and 5.1%. Those provided by the described Mathematical model obtained by Monte Carlo method are equal to 2.3 % in both samples. By comparison between the values obtained by the GUM uncertainty framework and a Monte Carlo method, the coverage interval obtained from the GUM framework in both samples is more conservative than that obtained by the MCM. A plausible explanation is that there are sources of uncertainty that weren’t taken into account and that influence the uncertainties associated with the estimates of the model parameters.

4. CONCLUSIONS

Thermal diffusivity and conductivity of uranium dioxide pellets samples, without and with addition of beryllium oxide were obtained by the flash method at 20 °C. The developed mathematical model performed an efficient and simultaneous implementation of corrections for all experimental problems associated to the flash method, encompassing evaluation of thermal conductivity and its assign uncertainties. Overall results indicate that exist a very good agreement between results obtained by the mathematical model and those measured by the original model of Parker. Both the GUM uncertainty framework and a Monte Carlo method were applied. However, it will always be necessary to make some assertions about
the uncertainties associated with the estimates of the model input quantities. The results also showed the increase at both quantities when it is added the beryllium oxide.

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REFERENCES


