Journal of Ceramic Processing Research. Vol. 17, No. 11, pp. 1132~1137 (2016)

J O U R N A L O F
Ceramic
Processing Research

Thermo-physical property review of uranium dioxide using IAEA online database THERPRO

Ji-Min Lee[a], Katsumi Yamada[b] and Yong-Soo Kim[a],*

[a]*Department of Nuclear Engineering, Hanyang University, 222, Wangsimni-ro, Seongdong-gu, Seoul 04763, Republic of Korea*
[b]*Section of Nucl. Power Technol. Development, Div. of Nucl. Power, Dept. of Nucl. Energy, Intern. Atomic Energy Agency, Vienna International Centre, 1400 Vienna, Austria*

The evaluation of reliable nuclear reactor performance under normal operation as well as under transient and accident conditions requires accurate representations of the thermo-physical properties of nuclear materials under high operating temperatures and in a harsh neutron irradiation environment. Thus, the thermo-physical properties of various reactor materials have been extensively studied since the 60's. Recently the IAEA collected, assessed, and stored more than 11,000 property data sets of about 1,600 materials in the THERPRO database running on their website. In this study, the thermal conductivity and heat capacity of uranium dioxide, the most widely used nuclear fuel material, are reviewed using the database, suggesting that the database can be usefully and powerfully utilized for nuclear material property research and development.

Key words: THERPRO, Nuclear materials, Uranium dioxide, Thermal conductivity, Heat capacity.

Introduction

Evaluation of fuel performance under normal operation as well as for accident analysis is vital for the safe design and operation of current and future nuclear reactor systems. This analysis always requires accurate representations of the thermo-physical properties of nuclear materials under high operating temperatures and in a harsh neutron irradiation environment. Particularly, since late 90's when high burn-up and extended fuel cycle operation was introduced, data on the material properties under such harsh environments have been demanded worldwide in the various fields of nuclear research and industry sectors.

Such widespread and international demand for detailed data has driven the IAEA (International Atomic Energy Agency) to conduct a collection and systemization of the data available in the public domain under the framework of the Coordinated Research Project (CRP) on the "Establishment of a Thermo-physical Properties Database for LWRs and HWRs" [1, 2]. One of the major outcomes was the web-based on-line THERPRO (THERmo-physical PROperties) database which provides registered users with thermo-physical material property data (Fig. 1) [3]. In total, seventeen kinds of material properties have been collected, including thermal conductivity, heat capacity, and thermal expansion coefficient. So far, more than 11,000 data sets of about 1,600 materials have been collected and compiled in the THERPRO database. Now recently published data in technical journals and reports are being continuously collected and added to the database.

Fig. 1. THERPRO DB homepage (http://therpro.iaea.org).

In this study, in order to demonstrate the usefulness and the power of the database, the thermo-physical property data of uranium dioxide, the most widely used nuclear fuel material, was chosen and reviewed using the THERPRO database.

In fact, it is well-known that the thermal conductivity of oxide fuel decreases substantially as the temperature increases. Thus, understanding the property changes of the fuel material is quite critical as it undergoes drastic temperature increases up to 2000 °C when operation begins. The conductivity reduction of operating fuel

*Corresponding author:
Tel : +82-2-2220-0467
Fax: +82-2-2281-5131
E-mail: yongskim@hanyang.ac.kr

considerably impoverishes heat transfer in the reactor core system, which leads to a loss of the operational safety margin.

On the other hand, a continued fission reaction increases fuel burn-up so the ceramic fuel experiences severe irradiation damage and irradiation effects with an enormous amount of fission products generated inside. This harsh irradiation environment inevitably deteriorates many material properties. This is the reason that the burn-up effect causing fuel property degradation has become one of the crucial concerns for advanced nuclear reactor development in the nuclear power industry. Therefore, burn-up dependent property data have been recently added to the THERPRO database.

Development of THERPRO Database

History of development

Since the early 90's, the IAEA has carried out two CRPs to collect data and systematize the thermo-physical property database of the materials used for light and heavy water nuclear reactors. The first project was carried out from 1990 to 1994 and collected data during the project period, and it was stored in a primitive electronic database format called THERSYST running on a DOS-based PC system [1] which was published in 1997. Later a large "spread" in some openly available data was found, so the Agency kicked off the second one to assess and to peer-review the property data, and to update the database with newly available data in the public domain [2].

In this 2nd CRP, nine institutes from seven countries participated: Atomic Energy of Canada Ltd (Canada); Nuclear Power Institute of China (China); University of West Bohemia (Czech Republic); Institute of Physics and Power Engineering and Institute of High Densities of Russian Academy of Science (Russian Federation); Bhabha Atomic Research Centre (India); Commissariat à I'Énergie Atomique (France); and Hanyang University and Seoul National University (Rep. of Korea). Significant contributions were also made by the Argonne National Laboratory (USA). Along with the project, the THERSYST database was completely redesigned and reconstructed using a powerful relational DBMS (Data Base Management System) using contemporary information technologies (O/S: Linux, DB: MySQL). The new web-based database used a GUI (Graphic User Interface) to enable data selection, manipulation, and representation in an easy and user-friendly manner. At the end of the meeting the new name THERPRO was given to this web-based database [3], and Hanyang university was designated by the IAEA as the responsible organization for the management and update of the database.

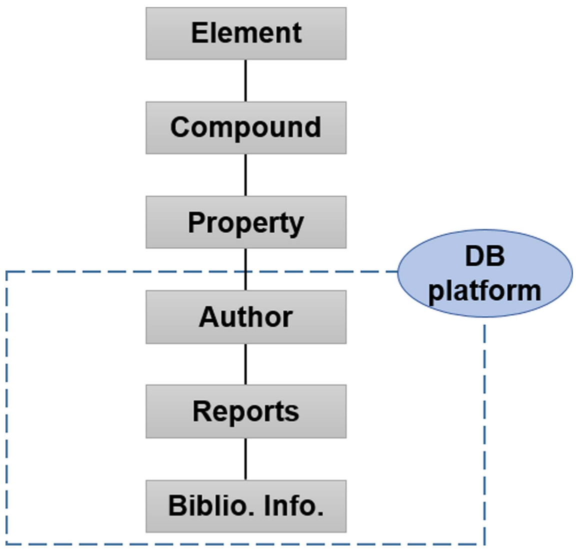


Fig. 2. Schematic structure of the THERPRO DB.

Structure of THERPRO database

The THERPRO DB has a hierarchical structure consisting of several levels, including the element, compound, property, and authors, as shown in Fig. 2. In actuality, in order to keep the data consistent and to facilitate easy comparison, all data are collected according to a standardized format consisting of five categories: material characterization, data characterization, measurement technique, bibliography, and numerical data. They are sorted and stored in the electric data platform of the DB management system, and all entries are interconnected using a network structure so that all data can be easily retrieved by an appropriate query action.

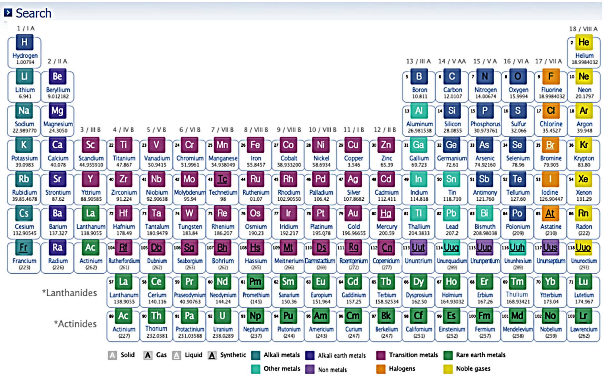
In practice, the database was developed as a common utility for engineers and researchers working in the nuclear laboratories and industries in all IAEA Member States. Nevertheless, the access to the source data files must be limited for database protection and for security purpose. Thus, every user is strongly recommended to register with identifiable information for authorization. Only authorized users can access the main structure of the database to obtain the bibliographic information.

The entire database system is currently running on the mainframe server of the Centre for Nuclear Materials Database (CNMD) at Hanyang university, Seoul, Korea. Its web address is http://therpro.iaea.org.

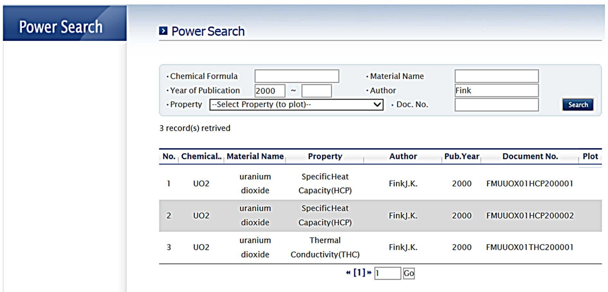
Data retrieval scheme of THERPRO

THERPRO provides three options for retrieving data: basic search, power search, and index search. Basic search is a step-by-step retrieval method from the web-site home page while the other two methods provide a more convenient and powerful data retrieval means.

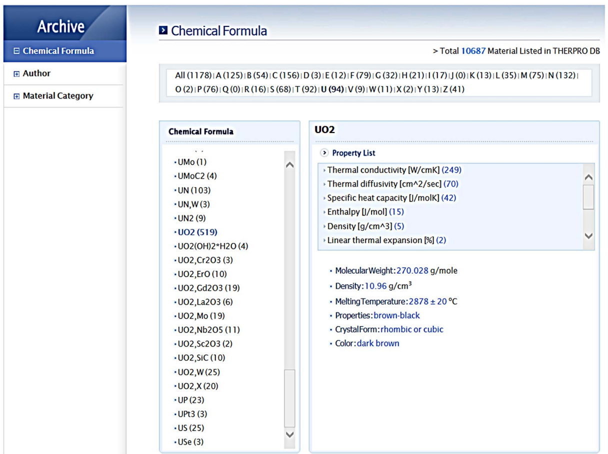
When you visit the web-site, visitor log-in is requested or strongly recommended. Once you enter the web site you will be led to the element level, the first entry layer to the main database. This was



(a) Top entry level in the basic search method



(b) An example of the power search method



(c) An example of the index search method (Archive)

Fig. 3. Three data retrieval schemes of the THERPRO DB.

designed with a periodic table interface to provide easy access to the target property data of the target material, as shown in Fig. 3(a).

Basic search begins with the clicking selection of the major element symbol of the target compound in the periodic table. It takes you to the next compound level, in which you will see the list of compound materials of the element. If you select one of the compounds in the list, you will be guided to the property level, which has the inventoried thermo-physical properties of the compound. The number in the parenthesis in this inventory list corresponds to the number of data sets collected and stored in the database. Upon choosing the property of your interest, you will be ushered to the database platform where all information on the author, report, and bibliography are correspondingly interconnected.

If you are familiar with THERPRO or know the author who published the target property of the compound of your interest, you can try the power search by selecting the menu button provided in the upper middle part of the home page (Fig. 3(b)). Simply typing the target compound and target property leads you directly to the database platform.

Since all property data collected in this database is indexed according to the chemical formula and compound name, you can also try the index search by selecting the ‘Archive’ menu on the home page (Fig. 3(c)), which also takes you to the database platform.

On reaching the DB platform, you will be requested to select the author from the author list as well as his or her report from the document list. Multiple choices of authors and their reports are possible. Appropriate selections and the ‘plot’ command will provide you with property data of interest with a graphic presentation.

As mentioned, every data set collected in this database has its own bibliographic information in a standardized format. An authorized user can access the file summarizing not only numerical data but also the details regarding data generation and corresponding measurement techniques.

Review of UO_2 Thermal Properties Using THERPRO Database

Thermal conductivity

As mentioned, the thermal conductivity of uranium dioxide follows the trend of typical ceramic materials. As the temperature rises, the conductivity decreases substantially and then increases steadily once the temperature enters the high temperature range. The former decrease is ascribed to the fact that the conductivity is based on phonon diffusion due to lattice vibrations, whereas the latter increase is owing to the contribution of free electrons in the high temperature regime.

Multiple selections of the data stored in the THERPRO DB, which were reported from the 1970’s to 2010’s, confirm the similar tendency of the temperature dependence in the graphic presentation of the search results (Fig. 4(a)). The figure shows that most of the measured data are in good agreement, with recent published data a little more conservative than previous measurements, whereas there is also a ‘spread’ in the high temperature measurements above 2000 K [4-13]. In fact, prior to the 90’s, there was a split in the opinion on whether the conductivity increase at high temperature is due to radiation heat transport or the free electron contribution. Later an

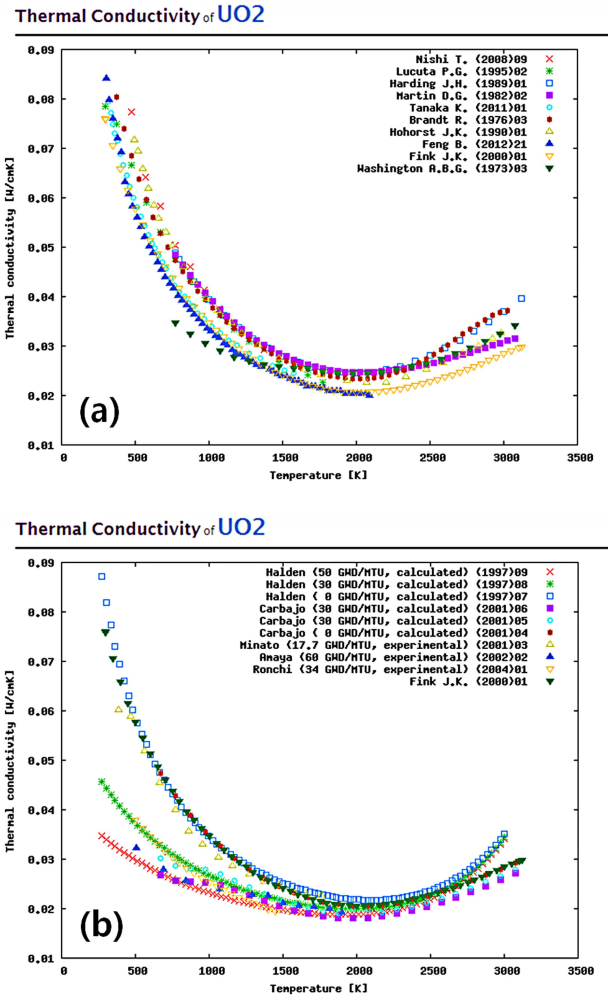


Fig. 4. Thermal conductivity of uranium dioxide (UO_2) (a) as a function of temperature and (b) as a function of temperature and burn-up.

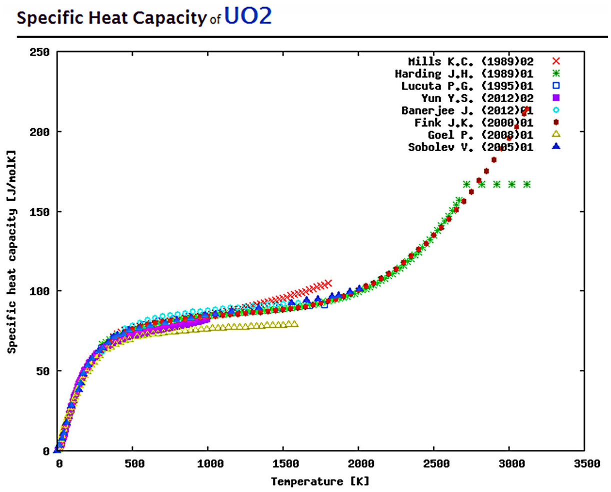


Fig. 5. Heat capacity of uranium dioxide (UO_2) as a function of temperature.

agreement was reached that it results from the free electron increase at high temperatures and their contribution to the heat transport. Fink [12] collected and reviewed all major experimental measurement data and models, and proposed following model based on the lattice phonon diffusion and electron contribution:

$$k = \frac{100}{7.5408 + 17.692t + 3.6142t^2} + \frac{6400}{t^{5/2}} \exp\left[\frac{-16.35}{t}\right] \quad (1)$$

where, $t = T(K)/1000$, and k is the thermal conductivity of 95% dense uranium dioxide in W m^{-1} K^{-1}. This study demonstrates that her model is a kind of best-estimated model of UO_2 thermal conductivity and justifies the reason why it is one of the most widely cited models.

On the other hand, the thermal conductivity is influenced by porosity, stoichiometric change, and fuel burn-up. In actuality, in order to buffer the fission gas swelling, an initially 95% TD (Theoretical Density) fuel pellet is loaded but the density change is insignificant during the reactor operation. The stoichiometry of the uranium dioxide is also maintained during operation, unless the fuel rod fails. Therefore, fuel burn-up is the primary factor that influences the conductivity degradation, principally due to irradiation damage.

In fact, the burn-up effect was anticipated even in the 80's but it was not explicitly reported until 1997. Based on the in-pile tests, Wiesenack [14] proposed the Halden model for 95% dense UO_2 in W m^{-1} K^{-1}:

$$k = \frac{1}{0.1148 + 0.0035B + 2.475 \times 10^{-4}(1 - 0.00333B)T} + 0.0132 exp(0.00188T) \quad (2)$$

where, T is the temperature in degrees Celsius, B is the fuel burn-up in MWd/kgU.

Since then, several researchers have carried out the investigation of thermal conductivity degradation experimentally or theoretically. Figure 4(b) shows recent research results [14-18] collected in the THERPRO database. As evident in the figure, there is still 'scatter' in the measurements, even though the Halden model seems to be a quite reliable model.

Specific heat capacity

One of the essential thermal properties of the ceramic nuclear fuel is its heat capacity because it determines the capability to keep the total generated heat in the reactor core, i.e., enthalpy which is one of the critical properties for reactor-safety analysis.

Figure 5 shows some of the recent data collected in the THERPRO database [12, 19-25]. Comparison of the data reveals that most of the reported data are fortunately in good agreement, even though they seem to be somewhat 'spread out' around the 1000 K range.

Because of its importance, the heat capacity of uranium dioxide has been studied by many researchers on a fundamental level since the 80's. The initial increase from room temperature to about 1000 K is ascribed to harmonic lattice vibrations, approximated by Debye theory. Between 1000 and 1500 K, the heat capacity increases slowly due to anharmonicity of the lattice vibrations caused by lattice thermal expansion. A rapid increase in heat capacity from 1500 to 2670 K arises from Frenkel type defects due to the generation of lattice and electronic defects. According to Harding [20], the heat capacity becomes saturated at 167.03 J mol^{-1} K^{-1} due to λ-phase transformation in the temperature range from 2670 to 3120 K, i.e., to the melting temperature. Above this transformation, the formation of Schottky type defects become dominant, whereas the Frenkel defects become saturated.

In the 2000's, Fink [12] also reviewed all available heat capacity data and models, and suggested her model of the heat capacity, C_p, in J mol^{-1} K^{-1} as a function of temperature as follows:

for 298.15 K $\leq T \leq$ 3120 K,

$$C_p = \frac{C_1 \theta^2 e^{\frac{\theta}{T}}}{T^2\left(e^{\frac{e}{T}} - 1\right)} + 2C_2T + \frac{C_3E_a e(-E_a/T)}{T^2} \quad (3)$$

where, $C_1 = 81.613$, $C_2 = 2.285 \times 10^{-3}$, $C_3 = 2.360 \times 10^{-7}$, $\theta = 548.68$, $E_a = 18531.7$, and T is the temperature in Kelvin.

In addition, she presented a polynomial equation as follow:

for 298.15 K $\leq T \leq$ 3120 K,

$$C_P = 52.1743 + 87.951t - 84.2411t^2 + 31.542t^3 - 2.6334t^4 - 0.71391t^{-2} \quad (4)$$

where, $t = T/1000$, and T is the temperature in Kelvin for a simple expression.

Currently, regardless of the heat capacity saturation due to λ-phase transformation, this polynomial equation has been widely used in reactor-safety calculations.

Conclusions

The international necessity to have accurate representations of thermo-physical material properties for safe design and operation of current and future nuclear reactor systems has driven the IAEA to conduct the collection and systemization of the data available in the public domain. One of the IAEA's recent activities in this area was to develop the THERPRO (THERmo-physical PROperties) database. It is a web-based on-line material properties database that provides registered users with assessed and peer-reviewed thermo-physical properties of nuclear materials. Currently, more than 11,000 property data of more than 1,600 materials have been stored in the database.

In this study, in order to demonstrate the usefulness and the power of the DB, thermal conductivity and heat capacity data of uranium dioxide were chosen and reviewed using the THERPRO database.

It was confirmed that the database has kept abundant essential data updated for the comparative analysis of data sets or from author to author. Furthermore, it is quite impressive that all data keeps its own bibliographic information summarizing not only numeric data but also the details on how the data was generated with what measurement techniques, along with the publication source.

In conclusion, therefore, utilization of the THERPRO database is highly recommended, as it is a reliable source of thermo-physical materials property data for various engineering and research applications according to a user's needs.

Acknowledgments

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MEST) (NRF-2014M2A7A1072569).

References

1. International Atomic Energy Agency, IAEA-TECDOC-949 (1997).
2. International Atomic Energy Agency, IAEA-TECDOC-1496 (2006).
3. S.D. Jo, S.B. Y Leon, J.S. Kim, and Y.S. Kim, J. Kor. Phys. Soc. 59 (2011) 1107-1110.
4. R. Brandt, G. Haufler, and G. Neuer, (Ed.: Y.S. Touloukian) CINDAS, Purdue Univ. (1976).
5. T. Nishi, A. Itoh, M. Takano, M. Numata, M. Akabori, Y. Arai, and K. Minato, J. Nucl. Mater. 376 (2008) 78-82.
6. B. Feng, A. Karahan, and M. S. Kazimi, J. Nucl. Mater. 427 (2012) 30-38.
7. J.K. Hohorst, NUREG/CR-5273 (1990).
8. P.G. Lucuta, Hj. Matzke, and R.A. Verrall, J. Nucl. Mater. 223 (1995) 51-60.
9. A.B.G. Washington, TRG-Report-2236(D) (1973) 1-61.
10. J.H. Harding and D.G. Martin, J. Nucl. Mater. 166 (1989) 223-226.
11. K. Tanaka, I. Sato, T. Hirosawa, K. Kurosaki, H. Muta, and S. Yamanaka, J. Nucl. Mater. 414 (2011) 316-319.
12. J.K. Fink, J. Nucl. Mater. 279 (2000) 1-18.
13. D.G. Martin, J. Nucl. Mater. 110 (1982) 73-94.
14. W. Wiesenack, in Proc. Int. Top. Mtg. on LWR Fuel Performance, Portland, Oregon, March 1997, p.507.
15. J.J. Carbajo, G.L. Yoder, S.G. Popov, and V.K. Ivanov, J. Nucl. Mater. 299 (2001) 181-198.
16. K. Minato, T. Shiratori, H. Serizawa, K. Hayashi, K. Une, K. Nogita, M. Hirai, and M. Amaya, J. Nucl. Mater. 288 (2001) 57-65.
17. M. Amaya, M. Hirai, H. Sakurai, K. Ito, M. Sasaki, T. Nomata, K. Kamimura, and R. Iwasaki, J. Nucl. Mater. 300 (2002) 57-64.
18. C. Ronchi, M. Sheindlin, D. Staicu, and M. Kinoshita, J. Nucl. Mater. 327 (2004) 58-76.
19. K.C. Mills, F.H. Ponsford, M.J. Richardson, N. Zaghini, and P. Fassina, Thermochim. Acta 139 (1989) 107-120.
20. J.H. Harding, D.G. Martin, and P.E. Potter, EUR 12402EN (1989) 1-90.

21. P.G. Lucuta, Hj. Matzke, and R.A. Verrall, J. Nucl. Mater. 223 (1995) 51-60.
22. Y.S. Yun, D. Legut, and P.M. Oppeneer, J. Nucl. Mater. 426 (2012) 109-114.
23. J. Banerjee, S.C. Parida, T.R.G. Kutty, A. Kumar, and S. Banerjee, J. Nucl. Mater. 427 (2012) 69-78.
24. P. Goel, N. Choudhury, and S.L. Chaplot, J. Nucl. Mater. 377 (2008) 438-443.
25. V. Sobolev, J. Nucl. Mater. 344 (2005) 198-205.