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# Uncertainty evaluation of the thermal expansion of simulated fuel

Chang Je Park\*, Kweon Ho Kang, Kee Chan Song

Korea Atomic Energy Research Institute, 150 Dukjin-dong, Yuseung-gu, Daejon 305-353, Republic of Korea

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#### Abstract

Thermal expansions of simulated fuel (SS1) are measured by using a dilatometer (DIL402C) from room temperature to 1900 K. The main procedure of an uncertainty evaluation was followed by the strategy of the  $UO_2$  fuel. There exist uncertainties in the measurement, which should be quantified based on statistics. Referring to the ISO (International Organization for Standardization) guide, the uncertainties of the thermal expansion are quantified in three parts—the initial length, the length variation, and the system calibration factor. Each part is divided into two types. The A type uncertainty is derived from the statistical iterative measurement of an uncertainty and the B type uncertainty comes from a non-statistical uncertainty including a calibration and test reports. For the uncertainty evaluation, the digital calipers had been calibrated by the KOLAS (Korea Laboratory Accreditation Scheme) to obtain not only the calibration values but also the type B uncertainty. The whole system, the dilatometer (DIL402C), is composed of many complex sub-systems and in fact it is difficult to consider all the uncertainties of sub-systems. Thus, a calibration of the system was performed with a standard material (Al<sub>2</sub>O<sub>3</sub>), which is provided by NETZSCH. From the above standard uncertainties, the combined standard uncertainties were calculated by using the law of a propagation of an uncertainty. Finally, the expanded uncertainty was calculated by using the effective degree of freedom and the *t*-distribution for a given confidence level. The uncertainty of the thermal expansion for a simulated fuel was also compared with those of UO<sub>2</sub> fuel.

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Keywords: Thermal expansion; Simulated fuel; Dilatometer

## 1. Introduction

Recently, plenty of studies on high burnup and high performance nuclear fuels have been performed by many researchers. However, these fuels are affected by a stress from the pellet cladding mechanical interaction (PCMI) and the fission gas for a long time and high temperature. Thus, to develop a new concept for fuels, it is necessary that fuel performance tests such as the material thermal, mechanical and chemical properties, irradiation behavior and a verification of the in-pile robustness tests are performed [1]. With the above fundamental data, many fuel performance code systems have been developed by using the thermal, mechanical, and chemical models of a nuclear fuel.

Thermal expansion as well as thermal conductivity is one of the most important thermophysical properties of a nuclear fuel. Most solid materials expand when heated up and shrink when cooled down. The thermal expansion is defined as a variation of the length with a temperature change, which is expressed as follows:

$$\Delta l/l_0 = (l_{\rm f} - l_0)/l_0 = \alpha_l (T_{\rm f} - T_0) \tag{1}$$

where  $l_0$  and  $l_f$  are the length at temperatures  $T_0$  and  $T_f$ , respectively and  $\alpha_l$  is the linear coefficient of a thermal expansion.

From the experiments, the linear coefficients of a thermal expansion for UO<sub>2</sub> fuel are distributed from  $1.03 \times 10^{-5}$  to  $1.08 \times 10^{-5}$  K<sup>-1</sup> [2].

In this study, a method to obtain the uncertainty of a thermal expansion of a simulated nuclear fuel [3] is suggested based on a previous approach. The weight percent of additives of the simulated fuel in this study are given in Table 1. The simulated fuel denotes a simulated irradiated fuel. When the fresh  $UO_2$  fuel loaded in the nuclear reactor, it is irradiated and the composition changes due to fission of the uranium. After a certain irradiation, the fuel contains various fission products, which are most toxic and radioactive. The prediction of thermal and mechanical properties of this irradiated fuel is very important for the fuel

<sup>\*</sup> Corresponding author at: Korea Atomic Energy Research Institute, Division of Dry Process Fuel Technology Development, 150 Dukjin-dong, Yuseung-gu, Daejon 305-353, Republic of Korea. Tel.: +82 42 868 2740; fax: +82 42 868 2403.

E-mail address: cjpark@kaeri.re.kr (C.J. Park).

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Table 1	
Weight percent of the additives of the simulated fuel	

Additive	Weight percent		
SrO	0.057		
Y <sub>2</sub> O <sub>3</sub>	0.04		
ZrO <sub>2</sub>	0.23		
La <sub>2</sub> O <sub>3</sub>	0.143		
CeO <sub>2</sub>	0.665		
Nd <sub>2</sub> O <sub>3</sub>	0.49		
Total	1.625		

 $SS1 = 98.375\% UO_2 + 1.625\%$  additives.

performance in the reactor. But, the high radioactivity prevents from ease treatment to measure properties. Thus, some toxic isotopes are replaced with non-toxic and stable isotopes when fabricating fuel pellet. This is a simulated fuel. The weights of impurities or additives are changed the irradiation period, which is called a burnup. The simulated fuel has been widely used to measure thermophysical and mechanical properties of an irradiated fuel instead. It is also known that both irradiated fuel and its simulated fuel exhibit similar behaviors of thermophysical and mechanical properties.

Until now, there have been several approaches to express the uncertainty of a thermal expansion. As a part work of obtaining the KOLAS (Korea Laboratory Accreditation Scheme) for a thermal expansion measurement of a nuclear fuel, a new approach for an uncertainty evaluation is developed based on the ISO (International Organization for Standardization) guide [4,5].

## 2. Uncertainty of the thermal expansion of nuclear fuel

Thermal expansions of simulated fuel are measured by using a horizontal type dilatometer (DIL402C, Netzsch) from room temperature to about 1900 K under an argon environment [6]. Table 2 shows the conditions for thermal expansion experiments in this study.

Table 2		
Conditions for thermal	expansion	experiment

Value
Dilatometer (DIL402C)
Room temperature $\sim$ 2273 K
Linear thermal expansion
5 K/min
2 points/K
$Al_2O_3$ (9.59 mm length)
Ar 99.9999%
Simulated fuel (SS1)
9.40 mm
95% theoretical density
95%
5
$293\pm10\mathrm{K}$
$14000 \text{ kg/m}^2$
4.2 ml/s
Alumina



Fig. 1. Thermal expansion and uncertainty for the reference material ( $Al_2O_3$ , NETZSCH).

The approach for an uncertainty of the thermal expansions of a simulated fuel starts from the following formulation based on the ISO guide [4,5],

$$f_{\rm E}(T) = (l(T) - l_0)/l_0 \times f_{\rm cal}$$
 (2)

where  $f_{\rm E}(T)$  is the thermal expansion, or output of experiments,  $l_0$  the sample length at the room temperature (mm), l(T) the sample length at  $T^{\circ}C$  (mm) and  $f_{\rm cal}$  is the ratio due to system calibration.

The factor of  $f_{cal}$ , which was introduced for the previous approach, has a unit value and its uncertainty comes from a system calibration test with the reference material [7]. If the results of the system calibration lie within a proper criterion, the system (DIL 402C) is thought to be a normal state and no other calibration is not performed. It contains two kinds of uncertainty: the first one is an iterative experiments uncertainty with a reference material  $(u_{f_{cal_1}}, type A)$  and the second one is a reference material uncertainty from a report ( $u_{f_{cal_2}}$ , type B). Fig. 1 shows a thermal expansion and its uncertainty for a certification report of a reference material, which was provided from NET-ZSCH. From Fig. 1, the uncertainty of the reference material from the certificate is very small and it does not really affect to the overall uncertainty of the thermal expansion experiment. The uncertainty of  $l_0$  is composed of three kinds: the first one comes from a resolution  $(u_{l_{01}}, \text{type B})$ , the second one comes from a calibration report ( $u_{l_{02}}$ , type B), and the third one from a variation of the room temperature ( $u_{l_{03}}$ , type B). The uncertainty of l(T)is derived from iterative experiments with a nuclear fuel  $(u_{lT},$ type A). From the above standard uncertainties, the combined standard uncertainties are calculated and the expanded uncertainty is calculated by the standard procedure for an uncertainty evaluation. Table 3 shows the uncertainty factors for the thermal expansion experiments.

The standard uncertainty of  $f_{cal}$  is obtained as

$$u_{f_{\text{cal}}}^2 = u_{f_{\text{cal}_1}}^2 + u_{f_{\text{cal}_2}}^2,\tag{3}$$

and the degree of freedom is derived as

$$\nu_{f_{\text{cal}}} = u_{f_{\text{cal}}}^4 / (u_{f_{\text{cal}_1}}^4 / \nu_{f_{\text{cal}_1}} + u_{f_{\text{cal}_2}}^4 / \nu_{f_{\text{cal}_2}}), \tag{4}$$

where  $v_{f_{cal_1}} = M - 1$ ,  $v_{f_{cal_2}} = (100/R)^2/2$ , *R* the degree of risk (=100 - confidence level, %) and *M* is the iterative experiment number.



Fig. 2. Flowchart of the uncertainty evaluation for the thermal expansion of the simulated fuel.

In this same way, the standard uncertainty of the initial sample length is calculated as

$$u_{l_0}^2 = u_{l_{01}}^2 + u_{l_{02}}^2 + u_{l_{03}}^2.$$
 (5)

The degree of freedom is set to be infinite from the guide.

Finally, the standard uncertainty of a length variation is given by the standard deviation such as

$$u_{l(T)}^{2} = s_{l}^{2}/N = 1/N \sum_{n=1}^{N} (l_{n} - \bar{l})^{2}/(N - 1),$$
(6)

where N is the iterative experiment number and  $l_n$  is the sample length for *n*th experiment (mm).

The degree of freedom is given as N - 1 in this case.

The combined uncertainty and its degree of freedom are obtained as

$$u_{\text{total}}^2 = C_{f_{\text{cal}}}^2 \cdot u_{f_{\text{cal}}}^2 + C_{l_0}^2 \cdot u_{l_0}^2 + C_{l(T)}^2 \cdot u_{l(T)}^2, \tag{7}$$

$$v_{\rm eff} = u_{\rm total}^4 / \left( u_{f_{\rm cal}}^4 / v_{f_{\rm cal}} + u_{l_0}^4 / v_{l_0} + u_{l(T)}^4 / v_{l(T)} \right)$$
(8)

and the sensitivity coefficients are obtained via a partial derivative of Eq. (1) as

$$C_{f_{\text{cal}}} = \partial f_{\text{E}} / \partial f_{\text{cal}} = (l(T) - l_0) / l_0, \qquad (9)$$

$$C_{l_0} = \partial f_{\rm E} / \partial l_0 = -f_{\rm cal} / l_0 ((l(T) - l_0) / l_0 + 1)$$
<sup>(10)</sup>

$$C_{l(T)} = \partial f_{\rm E} / \partial l(T) = f_{\rm cal} / l_0 \tag{11}$$

Table 3

Uncertainty parameters for the thermal expansion of a simulated fuel

Parameter	Type of uncertainty	d.f.
Initial length $(l_0)$		
Resolution $(l_{01})$	B (rectangular)	Inf.
Calibration $(l_{02})$	B (normal)	Inf.
Temperature variation $(l_{03})$	B (rectangular)	Inf.
Length $(l(T))$		
Iterative measurement	A (normal)	4
System calibration $(f_{cal})$		
Calibration test $(f_{cal_1})$	A (normal)	4
CRM report $(f_{cal_2})$	B (normal)	Inf.

The expanded uncertainty is obtained by multiplying the k-value from the Student's t distribution and the combined uncertainty. Fig. 2 shows the overall procedure of an uncertainty evaluation of the thermal expansion experiment of a simulated fuel. The approximate confidence level used in this study is 95% and the coverage factors (k) are obtained from the Table 4.

## 3. Results and discussions

Thermal expansion of a simulated fuel was performed by following the above procedure. Fig. 3 depicts the expanded uncertainty of the simulated fuel for various temperatures. And



Fig. 3. Thermal expansion and uncertainty for the simulated fuel as a function of the temperature.



Fig. 4. Comparison of the thermal expansion of the simulated fuel and  $\mathrm{UO}_2$  fuel.

Table 4 Uncertainty budget for the thermal expansion of a nuclear fuel (95% confidence level)					
Parameter	500 °C	500 °C			
	Standard uncertainty	Combined uncertainty	Standard uncertainty	Combined uncertainty	
Initial length, $l_0$ (mm)		$1.76 \times 10^{-3}$		$1.76 \times 10^{-3}$	

Parameter	500 °C		1000 °C		1500 °C	
	Standard uncertainty	Combined uncertainty	Standard uncertainty	Combined uncertainty	Standard uncertainty	Combined uncertainty
Initial length, $l_0$ (mm)		$1.76 \times 10^{-3}$		$1.76 \times 10^{-3}$		$1.76 \times 10^{-3}$
Resolution $(l_{01})$ Calibration $(l_{02})$ Temperature variation $(l_{03})$	0.001 $4.37 \times 10^{-4}$ $1.39 \times 10^{-3}$	(0.27) <sup>a</sup>	$\begin{array}{c} 0.001 \\ 4.37 \times 10^{-4} \\ 1.39 \times 10^{-3} \end{array}$	(0.19)	$\begin{array}{c} 0.001 \\ 4.37 \times 10^{-4} \\ 1.39 \times 10^{-3} \end{array}$	(0.16)
Length, <i>l</i> ( <i>T</i> ) (mm) Iterative measurement	$6.500 \times 10^{-4}$	$6.50 \times 10^{-4}$ (0.04)	$1.020 \times 10^{-3}$	$1.020 \times 10^{-3}$ (0.06)	$1.500 \times 10^{-3}$	$1.500 \times 10^{-3}$ (0.11)
System calibration ( $f_{cal}$ ) Calibration test ( $f_{cal_1}$ ) CRM report ( $f_{cal_2}$ )	$6.560 \times 10^{-2}$ $5.290 \times 10^{-3}$	$6.580 \times 10^{-2}$ (0.69)	$3.680 \times 10^{-2}$ $4.680 \times 10^{-3}$	$3.710 \times 10^{-2}$ (0.75)	$2.400 \times 10^{-2}$ $3.510 \times 10^{-3}$	$2.430 \times 10^{-2}$ (0.73)
Overall combined uncertainty $(u_{total})$	$3.660\times 10^{-4}$		$4.380\times10^{-4}$		$4.820\times10^{-4}$	

<sup>a</sup> Contribution factor (or importance factor) =  $C_i^2 u_i^2 / u_{\text{total}}^2$ .

Fig. 4 shows the thermal expansions of the simulated fuel and UO<sub>2</sub> fuel.

The thermal expansion of the simulated fuel is higher than that of UO<sub>2</sub>, which is due to the impurities in the simulated fuel. The behavior of thermal expansion less than 800 °C exhibits slightly higher thermal expansion of UO<sub>2</sub> but this could be seen as similar behavior and it could be neglected. But, the behavior of high temperature range from 800 °C, the difference of two values cannot be negligible any more because the errors of UO<sub>2</sub> and simulated fuels are different. In the previous experiments and discussion in Ref. [3], the higher thermal expansion for the simulated fuels indicate that the partial substitution of  $U^{4+}$  with (Y, La, and Nd)<sup>3+</sup> added in simulated fuel results in weakening the interatomic bonding in the solid solution matrix.

In Table 4, the uncertainties of the thermal expansions of the simulated fuel are given at the temperatures of 500, 1000, and 1500 °C, respectively. In this experiment, the thermal expansion data are obtained with 25 °C step. The three different temperatures in the results are chosen to see typical trend of uncertainty. The combined standard uncertainty of the initial length is  $1.755 \times 10^{-3}$  mm, which is the same for all the cases. The standard uncertainties of the iterative experiments of the length variation increase as the temperature increases. But the combined standard uncertainties of calibration decrease as temperature increases due to the behavior of the calibration report. In this table, contribution factors are defined as the ratio of the squares of the combined uncertainties for each factor. From the results, a system calibration is the most important factor, which affects the overall combined uncertainty for the temperature ranges. Initial length has a large effect on the uncertainty for a low temperature, but as the temperature increases the contribution of the uncertainty of the initial length slightly decreases. The overall expanded uncertainties slightly increase as the temperature increases, which are given in Table 5. The obtained coverage factors are about 2.0 for a 95% confidence. From the table, the expanded uncertainties of the thermal expansion of the simulated fuel increases as the temperature increases and the uncertainties were obtained as  $7.340 \times 10^{-4}$ ,  $8.791 \times 10^{-4}$ ,

Expanded uncertainty of the thermal expansion of the simulated fuel

Temperature (°C)	Thermal expansion	Expanded uncertainty	k
500	$4.64 \times 10^{-3}$	$7.34 \times 10^{-4}$	2.00
1000	$1.02 \times 10^{-2}$	$8.79  imes 10^{-4}$	2.01
1500	$1.70\times 10^{-2}$	$9.70  imes 10^{-4}$	2.01



Fig. 5. Contribution factors of the uncertainties.

and  $9.700 \times 10^{-4}$  for 500, 1000, and 1500 °C, respectively. The contribution factors of the uncertainties are depicted in Fig. 5. From Fig. 5, the contribution factors of components are changed as temperature changes and it was found that initial length is most dominant around low temperature range but the system calibration factor is most dominant for high temperature range. The contribution of sample length measurement keeps the lowest among three uncertainty factors.

#### 4. Conclusions and recommendations

Thermal expansion is measured with a dilatometer for a simulated fuel and its uncertainty is calculated as the temperature changes. There are three main uncertainty parameters including an initial length, the length at a temperature, and a system calibration with a reference material. In each part, a statistical standard uncertainty (type A) and a non-statistical standard uncertainty (type B) are combined to obtain an expanded uncertainty and an effective degree of freedom. From the results, the expanded uncertainty increases slightly as the temperature increases. And a system calibration is a major contributor to the overall expanded uncertainty in the thermal expansion experiments. As a conclusion, this study would be helpful in providing a fundamental approach to produce more reliable measurement data for the thermophysical properties of a nuclear fuel.

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### References

- D.R. Olander, Fundamental Aspects of Nuclear Reactor Fuel Elements, Technical Information Center, US Department Energy, 1976.
- [2] K.H. Kang, et al., J. Nucl. Mater. 301 (2002) 242.
- [3] K.H. Kang, et al., Int. J. Thermophys. 24 (2003) 1373.
- [4] ISO, Guide to the Expression of Uncertainty in Measurements, ISO, Geneva, 1993.
- [5] NIST, Guidelines for Evaluating and Expressing the Uncertainty of NIST Measurement Results, NIST Technical Note 1297, NIST, 1993.
- [6] Netzsch, Instruction Manual—High Temperature Dilatometer, DIL 402/C/7, Netzsch, 1997.
- [7] C.J. Park, et al., Proceedings of the First International Workshop on the Nuclear Fuel Thermophysical Property, Kyongju, Korea, November 8–9, 2004.