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MASTER

CODE DEVELOPMENT AND ANALYSIS PROGRAM

FUEL THERMAL EXPANSION (FTHEXP)

July 1978

EG&G Idaho, Inc.

IDAHO NATIONAL ENGINEERING LABORATORY

DEPARTMENT OF ENERGY

IDAHO OPERATIONS OFFICE UNDER CONTRACT EY-76-C-07-1570

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PRELIMINARY ANALYSIS REPORT

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FUEL THERMAL EXPANSION (FTHEXP)

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CONTENTS

FOREWORD	1
4.0 FUEL THERMAL EXPANSION	2
4.1 Summary	2
4.2 Review of Literature	4
4.3 Model Development	5
4.3.1 Low Temperature Thermal Expansion	5
4.3.2 High Temperature Thermal Expansion	5
4.3.3 Mixed Oxide Thermal Expansion	7
4.3.4 Thermal Expansion of Partially Molten Fuel	7
4.3.5 Thermal Expansion of Entirely Molten Fuel	8
4.4 Model-Data Comparison and Uncertainty	8
4.5 Listing of the FTHEXP Subcode	12
4.6 References	16

LIST OF FIGURES

A-4.1 Correlation for the Thermal Expansion Strain of UO ₂ Compared With Its Data Base	9
A-4.2 Correlation for the Thermal Expansion Strain of PuO ₂ Compared With Its Data Base	10
A-4.3 Comparison of the UO ₂ , PuO ₂ , and (U _{0.8} , Pu _{0.2})O ₂ Correlations From 0 to 2000 K	11
A-4.4 Comparison of the Predictions of This Model and Those of the Model in MATPRO-10 ^[A-4.15] for UO ₂	13

LIST OF TABLES

A-4.I Parameters Used in UO ₂ and PuO ₂ Solid Phase Thermal Expansion Correlations	3
A-4.II Listing of the Subcode FTHEXP	14

FOREWORD

This report describes work which is part of the fuel rod behavior modeling task performed at EG&G Idaho, Inc. It is an interim addition to work previously published in the Materials Properties (MATPRO) Handbook^[a] and will replace Section A-4 (Fuel Thermal Expansion) of the MATPRO 11 handbook. This update of the fuel thermal expansion subcode incorporates new data and includes an estimate of the error in the calculated strains.

The material property correlations and computer subcodes described in MATPRO are developed for use in Light Water Reactor (LWR) analytical programs such as the Fuel Rod Analysis Program -- Steady State (FRAP-S3)^[b] code and the Fuel Rod Analysis Program -- Transient (FRAP-T4)^[c] code. This work is being performed as part of a broad effort to develop and verify analytical models capable of describing nuclear fuel rod behavior.

The format and numbering scheme used in this report is consistent with its intended use as the fourth section of Appendix A of the MATPRO handbook. It is beyond the scope of this report to provide a complete description of the MATPRO subcode and its organization. Readers who require such a description should consult the MATPRO 10 handbook^[a]. Readers who require descriptions of the use of materials properties subcodes should consult the code descriptions^[b, c].

[a] G. A. Reymann (Ed.) MATPRO Version 10 - A Handbook of Materials Properties for Use in the Analysis of Light Water Reactor Fuel Rod Behavior, TREE-NUREG-1180 (February 1978).

[b] J. A. Dearien, et al., FRAP-S3: A Computer Code for Steady-State Analysis of Oxide Fuel Rods - Vol 1 - Analytical Models and Input Manual, TFBP-TR-164, Revision 2 (March 1978).

[c] J. A. Dearien, et al., FRAP-T4: A Computer Code for Transient Analysis of Oxide Fuel Rods - Vol 1 - Analytical Models and Input Manual, CDAP-TR-78-027 (July 1978).

4. FUEL THERMAL EXPANSION (FTHEXP)

A model is presented in this report which deals with dimensional changes in fuel pellets caused by changes in temperature. It is capable of dealing with any combination of UO_2 and PuO_2 in solid, liquid or mixed phase states, and includes expansion due to the solid-liquid phase change.

Fuel dimensional changes affect the pellet-to-cladding gap size, which is a major factor in determining gap heat transfer and thus the stored energy, an important quantity for safety analysis.

4.1 SUMMARY

The function FTHEXP models fuel thermal expansion as a function of temperature, fraction of PuO_2 , and the fraction of fuel which is molten. The oxygen to metal ratio (O/M) is not included. When the departure from stoichiometry, $|O/M-2.0|$, is greater than 0.2, there is clearly an effect [A-4.1, 4.2, 4.3]. However typical reactor fuels only deviate about a tenth this much from the stoichiometric composition.

The equations for the thermal expansion of UO_2 and PuO_2 have the same form. In the solid phase the equation is

$$\frac{\Delta L}{L_0} = K_1 T - K_2 + K_3 \exp(-E_D/kT) \quad (A-4.1)$$

where

$\frac{\Delta L}{L_0}$ = linear strain caused by thermal expansion
(equal to zero at 300 K) (unitless)

T = temperature (K)

k = Boltzmann's constant (1.38×10^{-23} J/K)

and the other parameters are given in Table A-4.I.

TABLE A-4.1

PARAMETERS USED IN UO_2 AND PuO_2 SOLID PHASE
THERMAL EXPANSION CORRELATIONS

Constant	UO_2	PuO_2	Units
K_1	1.0×10^{-5}	9.0×10^{-6}	K^{-1}
K_2	3.0×10^{-3}	2.7×10^{-3}	Unitless
K_3	4.0×10^{-2}	7.0×10^{-2}	Unitless
E_D	6.9×10^{-20}	7.0×10^{-20}	J

For mixed UO_2 and PuO_2 , the thermal expansion of the solid is found by combining the contribution from each constituent in proportion to its weight fraction.

During melting an expansion equal to a linear strain of 0.043 occurs. If the fuel is partially molten, the strain due to thermal expansion is given by

$$\frac{\Delta L}{L_0} = \frac{\Delta L}{L_0} (T_m) + 0.043 \cdot \text{FACMOT} \quad (\text{A-4.2})$$

where

$$\frac{\Delta L}{L_0} (T_m) = \text{thermal expansion strain of solid fuel from Equation (A-4.1)} \\ \text{with } T = T_m$$

$$T_m = \text{melting temperature of the fuel (K)}$$

FACMOT = fraction of the fuel which is molten (unitless)

FACMOT = 0.0 - fuel all solid

FACMOT = 1.0 - fuel all molten.

The correlation used to describe the expansion of entirely molten fuel is

$$\frac{\Delta L}{L_0} = \frac{\Delta L}{L_0} (T_m) + 0.043 + 3.6 \times 10^{-5} [T - (T_m + \Delta T_m)] \quad (\text{A-4.3})$$

The solid to liquid phase transition is isothermal only for pure UO_2 or pure PuO_2 . For $(\text{U}, \text{Pu})\text{O}_2$ the transition occurs over a finite temperature range, denoted in Equation (A-4.3) by ΔT_m .

The uncertainty of the pooled data was found to be temperature dependent, increasing approximately linearly with temperature. Therefore a percentage error is given rather than a fixed number. The 2σ limits were found to be $\pm 10\%$ of the calculated value.

Section 4.2 contains a discussion and evaluation of the sources used. Section 4.3 presents the development of the model. In Section 4.4 the model predictions are compared with data and an uncertainty estimate is given. Sections 4.5 and 4.6 give listings of the FORTRAN subcode FTHEXP and the references.

4.2 Review of Literature

Data were taken from nine sources for UO_2 [A-4.1-4.9] and two sources for PuO_2 [A-4.3, 4.12]. For UO_2 the data cover a temperature range from 300 to 3400 K, and for PuO_2 from 300 to 1700 K.

In four of the UO_2 experiments [A-4.1, 4.2, 4.8, 4.9], X-ray measuring techniques were used. This type of measurement gives the change in the lattice parameter rather than the bulk thermal expansion. Several investigators [A-4.2, 4.10, 4.11] have noted that the change in the lattice parameter is appreciably smaller than the bulk thermal expansion measured using dilatometric or interferometric methods, especially at high (> 1000 K) temperature. In general the difference is attributed to the creation of Schottky defects [A-4.2, 10, 11]. Hoch and Momin [A-4.9] obtained results where there was no discrepancy between their X-ray results and bulk results. However, the bulk of the data support the Schottky defect idea since the X-ray data consistently fall below the others at high temperatures, where defects begin to appear in large numbers. Therefore X-ray data were used only at lower temperatures (< 800 K).

4.3 Model Development

While most authors simply fit their data with a polynomial, in this paper correlations based on more physical grounds are used.

4.3.1 Low Temperature Thermal Expansion. The simplest theory of the linear expansion of a solid near room temperature is found in most elementary physics texts such as Sears and Zemansky^[A-4.13].

$$\Delta L = L_0 K_1 (T - T_0) \quad (A-4.4a)$$

$$\text{or } \frac{\Delta L}{L_0} = K_1 T - K_1 T_0 \quad (A-4.4b)$$

where

K_1 = the average coefficient of linear expansion (K^{-1})

T_0 = a reference temperature (K)

L_0 = length at reference temperature (m)

at the reference temperature $\Delta L=0$ or equivalently $L=L_0$.

The low temperature (< 800 K) data were fit by the method of least squares to a generalized form of Equation (A-4.4b).

$$\frac{\Delta L}{L_0} = K_1 T - K_2 \quad (A-4.4c)$$

This fit was done separately for UO_2 and PuO_2 and the coefficients K_1 and K_2 for each material are listed in Table A-4.I. The numbers in the table have been rounded off to two significant figures. Comparing Equations (A-4.4b) and (A-4.4c) shows that $T_0 = K_2/K_1$, which for both fuels is 300 K, a temperature typical of the reference temperatures in the data bases. These correlations describe low temperature thermal expansion within the data scatter.

4.3.2 High Temperature Thermal Expansion. For both UO_2 and PuO_2 Equation (A-4.4c) was inadequate at higher temperatures ($T > 1000$ K). This is most likely due to the formation of Schottky defects. Frenkel defects will also be present, but should have no measurable effect on the thermal expansion^[A-4.2, A-4.9]. The contribution from Schottky defects should be directly proportional to their concentration^[A-4.2] which is given by^[A-4.2, 4.14]

$$\frac{N}{N_0} = K_3 \exp (-E_D/kT) \quad (A-4.5)$$

where

N = number of Schottky defects in the crystal

N_0 = number of atoms in the crystal

E_D = energy of formation of a defect (J)

k = Boltzmann's constant (1.38×10^{-23} J/K)

K_3 = constant to be determined (unitless)

The difference between the thermal strain calculated with Equation (A-4.4c) and each data point was found. These differences were assumed to be the defect contributions to the thermal expansion strain and were fit by the method of least squares to an equation of the form

$$\left(\frac{\Delta L}{L_0}\right)_D = K_3 \exp (-E_D/kT) \quad (A-4.6)$$

where

$$\left(\frac{\Delta L}{L_0}\right)_D = \text{defect contribution to the thermal expansion (unitless).}$$

The values for K_3 and E_D resulting from these fits are given in Table A-4.I.

Baldock et al^[A-4.2] did a similar analysis using their UO_2 data and those of Conway et al^[A-4.4]. Both the pre-exponential factor, K_3 , and the energy of formation, E_D , were larger than those listed in Table A-4.I. These differences mean that Baldock's Schottky term is smaller than the one found here at low temperatures and larger at high temperatures. The magnitude of the Schottky term determined this way is strongly dependent on the low temperature correlation used. Since Equation (A-4.4c) has been found using a much broader data base than Baldock's, the values for K_3 and E_D in Table A-4.I should be the more accurate, and are the ones used in this model.

4.3.3 Mixed Oxide Thermal Expansion. When the fuel is composed of a mixture of UO_2 and PuO_2 , the thermal expansion is found by taking a weighted average of the contributions from each component:

$$\left(\frac{\Delta L}{L_0}\right)_{(U,Pu)O_2} = \left(\frac{\Delta L}{L_0}\right)_{UO_2} * (1.0 - COMP) + \left(\frac{\Delta L}{L_0}\right)_{PuO_2} * COMP \quad (A-4.7)$$

where

COMP = PuO_2 weight fraction.

4.3.4 Thermal Expansion of Partially Molten Fuel. Christensen^[A-4.6] has determined that UO_2 experiences a linear thermal strain of 0.043 on melting. His measurements show considerable scatter but are the only data available. There are no comparable measurements for PuO_2 . The structure of the two fuels is similar enough however, so that no serious error should be introduced by equating the PuO_2 expansion on melting with that of UO_2 . For partially molten fuel the thermal expansion strain is given by

$$\frac{\Delta L}{L_0} = \frac{\Delta L}{L_0} (T_m) + 0.043 * FACMOT \quad (A-4.8)$$

The various terms of Equation (A-4.8) are defined in Section 4.1.

4.3.5 Thermal Expansion of Entirely Molten Fuel. The experiment of Christensen on UO_2 again produced the only data available and must be used for all combinations of $(\text{U}, \text{Pu})\text{O}_2$.

A least squares fit to his limited data yields

$$\frac{\Delta L}{L_0} = \frac{\Delta L}{L_0} (T_m) + 0.043 + 3.6 \times 10^{-5} [T - (T_m + \Delta T_m)] \quad (\text{A-4.9})$$

where the variables have all been previously defined.

4.4 Model-Data Comparison and Uncertainty

Figure A-4.1 shows a comparison of the correlation for UO_2 and its data base. The three very low points around 1500 K are all from Christensen^[A-4.6]. Other data from him fit well to the curve and there is no obvious reason for the large deviation of these points. At the highest temperatures there are several data considerably above the curve. These are also from Christensen. At these temperatures there is a possibility that the fuel was partially molten due to an uneven temperature distribution in the sample. The large expansion which occurs on melting could easily explain the deviation of these data from the solid UO_2 data.

A similar comparison of the correlation and the data is shown in Figure A-4.2 for PuO_2 . Figure A-4.3 shows a comparison of the expansion curves for UO_2 and PuO_2 , and $(\text{U}_{0.8}, \text{Pu}_{0.2})_2$. No data are shown on this curve because thermal expansion data for mixed oxides are not available. The figures show that the thermal expansion behavior of the two materials differ, but only slightly.

Error bands, calculated from the sum of the squared residuals, are shown in Figures A-4.1 and A-4.2 as dotted lines. These reflect a standard error of $\pm 10\%$ of the calculated value found from the UO_2 data set. A percentage uncertainty is given because the error increases with temperature. A single valued uncertainty can lead to a non-physical possibility in this model. For example, the standard error for UO_2 is $\pm .0012$, which equals the thermal expansion strain at 420 K. Thus for any temperature less than 420 K,

Fig. A-4.1 Correlation for the Thermal Expansion Strain of UO_2 compared with its data base.

Fig. A-4.2 Correlation for the Thermal Expansion Strain of PuO₂ compared with its data base.

Fig. A-4.3 Comparison of the UO_2 , PuO_2 , and $(\text{U}_{0.8}, \text{Pu}_{0.2})\text{O}_2$ correlations from 0 to 2000 K.

the lower limit implied by the uncertainty would be negative, implying that as the fuel heats from 300 to 400 K, it contracts. A percentage error automatically precludes this.

The error for PuO_2 was somewhat smaller, but this is probably due to the limited number of sources. The $\pm 10\%$ error limit is also used for PuO_2 to avoid assigning unrealistic accuracy to these data.

4.5 Listing of the FTHEXP Subcode

A FORTRAN listing of the FTHEXP subcode is given in Table A-4.II. The estimated standard error is defined within the subcode but is not returned. In a further simultaneous revision of FTHEXP and the INEL fuel rod analysis codes which use it, all uncertainties will be stored in a common block to facilitate uncertainty studies.

The code presented here is considerably different from that found in the previous fuel thermal expansion subcode^[A-4.15]. However when the results of each code are plotted, as is done in Figure A-4.4, it is evident that the predictions are close for low temperatures. The primary difference occurs above the melting point where this code predicts both a larger increase on melting and more expansion after melting than did the previous one. These differences are due primarily to a re-analysis of Christensen's data. Since it is based on physical principles, the new model may be extrapolated beyond its data base with more confidence than the previous one. This is especially important at the higher temperatures, where the two models differ the most.

Figure A-4.4 Comparison of the predictions of this model and those of the Model in MATPRO-10[A-4.15] for UO_2 .

TABLE A-4.II

PARAMETERS USED IN UO_2 and PuO_2
SOLID PHASE THERMAL EXPANSION CORRELATIONS

FUNCTION FTHEXP (FTEMP,FACMOT)

THE SUBPROGRAM FTHEXP IS USED TO CALCULATE THE STRAIN OF THE FUEL CAUSED BY THERMAL EXPANSION. THERMAL EXPANSION STRAIN IS CALCULATED FOR UO_2 , PuO_2 , OR $(U,Pu)O_2$ AS A FUNCTION OF TEMPERATURE, PuO_2 CONTENT, AND FRACTION OF THE FUEL WHICH IS MOLTEN. THE CORRELATIONS WERE DERIVED WITH THE ASSUMPTION THAT THE THERMAL EXPANSION STRAIN IS ZERO AT 300K.

FTHEXP = OUTPUT FUEL STRAIN DUE TO THERMAL EXPANSION (UNITLESS)
UFTHEX = ESTIMATED STANDARD ERROR OF FTHEXP (PER CENT)
(NOT CURRENTLY RETURNED).

FTEMP = INPUT FUEL TEMPERATURE (K)
FACMOT = INPUT FUEL FRACTION WHICH IS MOLTEN (UNITLESS)
FACMOT = 1.0 - FUEL IS ALL MOLTEN
FACMOT = 0.0 - FUEL IS ALL SOLID

THE CORRELATIONS USED IN THIS SUBROUTINE ARE BASED ON DATA FROM
(1) P.J.BALDOCK ET AL, JOURNAL OF NUCLEAR MATERIALS, 16 (1966)
(2) N.H.BRETT AND L.E.RUSSEL, PROCEEDINGS OF THE 2ND INTERNATIONAL CONFERENCE ON PLUTONIUM METALLURGY, GRENOBLE, FRANCE (1960) PP.397-410.
(3) M.D.BURDOCK AND H.S.PARKER, JOURNAL OF THE AMERICAN CERAMIC SOCIETY, 39 (1956) PP. 181-187.
(4) J.A.CHRISTENSEN, JOURNAL OF THE AMERICAN CERAMIC SOCIETY, 46 (1963) PP. 607-608.
(5) J.B.CONWAY ET AL, TRANSACTIONS OF THE AMERICAN NUCLEAR SOCIETY, 6 (1963).
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(10) M.TOKAR ET AL, NUCLEAR TECHNOLOGY, 17 (1972) PP.147-152.

FTHEXP WAS ORIGINALLY CODED BY V.F.BASTON IN MARCH 1974
MODIFIED BY C.S.GILSEN IN FEB. 1975
LAST MODIFIED BY G.A.REYMANN IN JULY 1978

COMMON / PHYPRC / FTMELT,FHEFUS,CTMELT,CHEFUS,CTRANB,
CTRANE,CTRANZ,FDELTA,BU,COMP

THE PHYPRC COMMON BLOCK IS SET BY CALL TO THE SUBROUTINE PHYPRO WHICH IS PART OF THE MATERIALS PROPERTIES PACKAGE USED AT THE INEL.

QUANTITIES CONTAINED IN IT USED IN THIS SUBCODE ARE

COMP = PuO_2 CONTENT (WT. %)

FTMELT = FUEL MELTING TEMPERATURE (K)

FDELTA = LIQUID-SOLID COEXISTENCE TEMPERATURE RANGE (K)

WHEN THE FUNCTION FTHEXP IS USED ALONE, NOT AS PART OF THE INEL CODE, VALUES FOR COMP, FTMELT, AND FDELTA MUST BE INPUT.

DATA C1U,C2U,C3U,EDU / 1.0E-05, 3.0E-03, 4.0E-02, 6.9E-20 /
DATA C1PU,C2PU,C3PU,EDPU / 9.0E-06, 2.7E-03, 7.0E-02, 7.0E-20 /

FTX (C1,C2,C3,ED,BK,T) = $C1 \cdot T - C2 + C3 \cdot \exp(-ED/(BK \cdot T))$

T = FTEMP

TABLE A-4.II

LISTING OF THE SBUCODE FTHEXP
(continued)

```

C      BK      = 1.38E-23
C      BK      = BOLTZMANN'S CONSTANT (J/K)
C      COMP    = COMP/100.0
C      IF(T .GT. FTMELT) GO TO 25
C      FTHEXP = FTX(C1U ,C2U ,C3U ,EDU ,BK,T)*(1.0 - COMP) +
C              FTX(C1PU,C2PU,C3PU,EDPU,BK,T)*COMP
C      GO TO 100
C 25  FTHEXM = FTX(C1U ,C2U ,C3U ,EDU ,BK,FTMELT)*(1.0 - COMP) +
C      FTX(C1PU,C2PU,C3PU,EDPU,BK,FTMELT)*(COMP)
C      IF(T .GE. (FTMELT+FDELTA)) GO TO 75
C      FTHEXP = FTHEXM + .043*FACMOT
C      GO TO 100
C 75  FTHEXP = FTHEXM + .043 + 3.6E-05*(T-(FTMELT+FDELTA))
C 100 CONTINUE
C      UFTHEX = 10.0
C      RETURN
C      END

```

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