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FUEL VISCOSITY (FVISCO)

C. S. Watson
D. L. Hagrman

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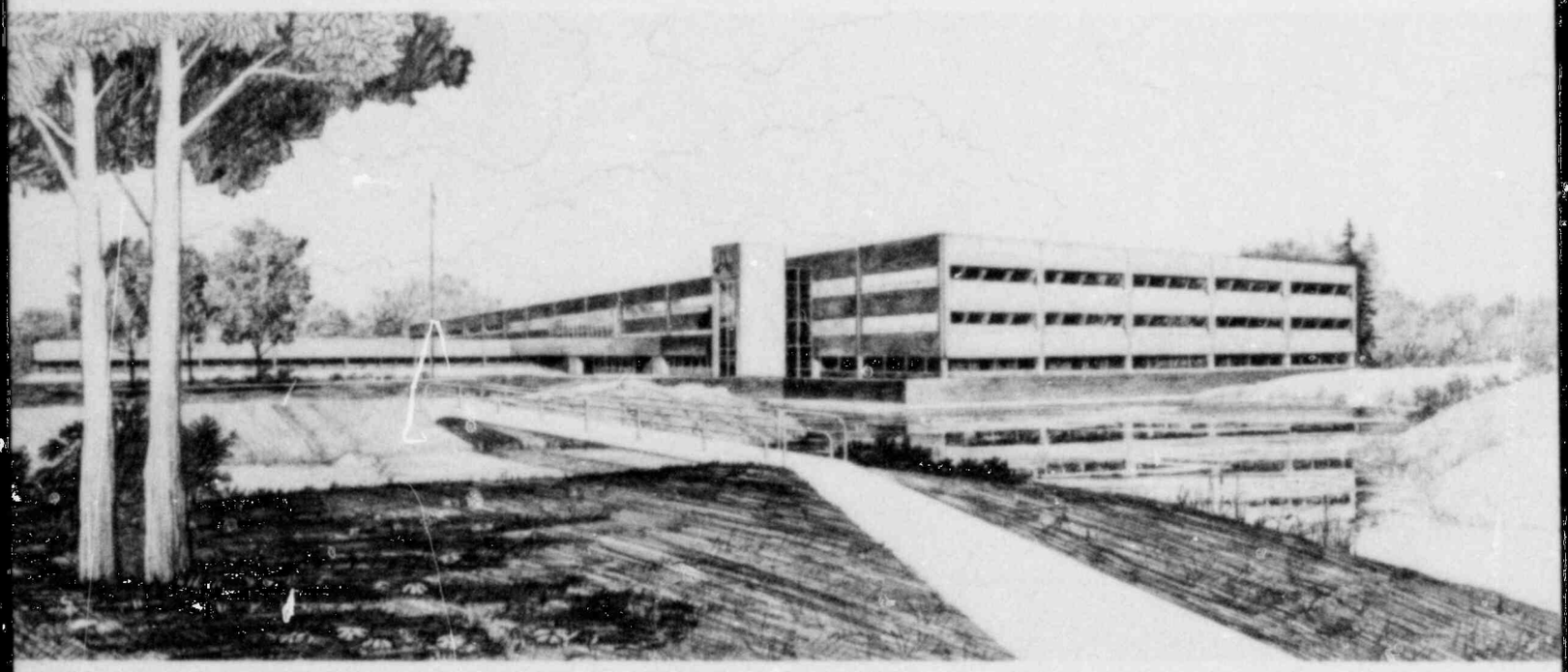

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Author(s): C. S. Watson
D. L. Hagrman

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EG&G Idaho, Inc.
Idaho Falls, Idaho 83415

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CONTENTS

LIST OF FIGURES

LIST OF TABLES

FOREWORD

This report describes a model for the viscosity of uranium dioxide as a function of temperature and fraction of molten fuel. The report will become part of an update to the Materials Properties (MATPRO) Handbook[a] used in the fuel rod behavior modeling task performed at the INEL.

A viscosity model for uranium dioxide was developed to provide a preliminary description of molten fuel viscosity and to define data needs. The model is preliminary because the effects of departure from stoichiometry and the range of temperatures where liquid and solid UO_2 can coexist are not modeled. Also the model does not consider any possible contamination of the molten uranium dioxide. Uncertainty estimates are provided based on the data used in the model.

The format and numbering scheme used in this report are consistent with its intended use as an update to the MATPRO handbook. Readers who require descriptions of the rest of the material properties package or of the use of this package should consult the code descriptions[a,b,c].

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a D. L. Hagrman, G. A. Reymann, R. E. Mason, MATPRO-Version 11 Revision 1: A Handbook of Materials Properties for Use in the Analysis of Light Water Reactor Fuel Rod Behavior, NUREG/CR-0497 TREE-1280, Rev 1, (February 1980).

b G. A. Berna et al., FRAPCON-1: A Code for the Steady-State Analysis of Oxide Fuel Rods, CDAP-TR-78-032-R1, (November 1978).

c L. J. Siefken et al., FRAP-T5: A Computer Code for the Transient Analysis of Oxide Fuel Rods, NUREG/CR-0840 TREE-1281, (June 1979).

15. FUEL VISCOSITY (FVISCO)

(C. S. Watson and D. L. Hagrman)

The function FVISCO calculates the dynamic viscosity of uranium dioxide. The viscosity is one of the parameters needed to model the motion of fuel during severe core damage.

15.1 Summary

The viscosity of uranium dioxide is modeled as a function of temperature, melting temperature (solidus), and the fraction of the fuel that has liquified. Input arguments describing the oxygen-to-metal ratio and plutonium dioxide content are not used in the current correlations for viscosity.

Viscosity is calculated by one of three equations, depending on whether the temperature is below the melting point for uranium dioxide, in the range of temperatures where liquid and solid uranium dioxide can coexist, or above this range.

The equation used to model the viscosity of completely liquified fuels is

$$\eta_\ell = 1.23 \times 10^{-2} - 2.09 \times 10^{-6} \, T \qquad \text{(A-15.1)}$$

where

η_ℓ = dynamic viscosity of the liquid (Pa s).

T = temperature (K).

For solid uranium dioxide the viscosity is modeled with the expression

$$\eta_s = 1.38 \exp(4.942 \times 10^4/T) \qquad \text{(A-15.2)}$$

where

η_s = dynamic viscosity of the uranium dioxide for temperatures below melting (Pa s).

In the temperature range where liquid and solid uranium dioxide phases can both exist, the viscosity is modeled with the expression

$$\eta = \eta_s (1.-f) + \eta_\ell f \qquad \text{(A-15.3)}$$

where

η = dynamic viscosity of the liquid-solid mixture (Pa s)

f = fuel fraction that is liquid (unitless).

The estimated uncertainty of the values computed with Equations (A-15.1) to (A-15.3) is computed with the FVISCO subcode but not returned as an output argument. The expressions used for this uncertainty are

$$U = \eta A (1 + |Y - 2|) \qquad \text{(A-51.4)}$$

where

U = estimated uncertainty (Pa s)

A = 0.33 for temperatures above melting
0.67 for temperatures below melting

Y = oxygen-to-metal ratio of the fuel (unitless).

Details of the development of the fuel viscosity model used in the FVISCO function are presented in the following sections. Section 15.2 is a review of the data and Section 15.3 is a discussion of the model development. The subcode is listed in Section 15.4 and references are provided in Section 15.5.

15.2 Fuel Viscosity Data

Viscosities for solid UO_2, $UO_{2.06}$, and $UO_{2.16}$ have been reported by Scott, Hall and Williams[A-15.1] Viscosities for the non-stoichiometric oxides are lower than the viscosity of UO_2 at corresponding temperatures and thus could be measured over a sufficient range to establish the following relation for non-stoichiometric UO_2

$$\eta_s = A \exp(-B/T) \qquad \text{(A-15.5)}$$

where A and B are material constants and η_s and T were defined in the summary. The viscosity of UO_2 was determined to be 2×10^{11} Pa s at 1923 K and to be in excess of 10^{17} Pa s at 1273 K.

Viscosity data at much higher temperatures were obtained by Nelson[A-15.2, A-15.3]. An early measurement (0.145 Pa s at a temperature of 3028 K) was reported to correspond to incomplete melting of the sample. Subsequent data (0.045 Pa s at 3028 K and 0.036 at 3068 K) represent a viscous fluid at temperatures below the melt temperature used in the MATPRO Handbook[a]. These data are not suitable for use in the viscosity model because all three measurements have indicated viscosities well above the more extensive measurements at temperatures where the uranium dioxide is known to be completely liquified.

Two useful sources of data with completely molten uranium dioxide are available. Tsai and Olander[A-15.4] published data from two samples and Woodley[A-15.5] published more extensive data from a single sample. The data are tabulated in Tables A-15.I and A-15.II and plotted in Figure A-15.1. The precision of the data by Woodley is noticeably higher than the precision of the other data, but there is a larger difference between the two

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[a] The melt temperature for UO_2 is given as 3113 K in the PHYPRP subcode of the MATPRO package. This package is described in D. L. Hagrman, G. A. Reymann, and R. E. Mason, MATPRO-Version 11 Revision 1: A Handbook of Materials Properties for Use in the Analysis of Light Water Reactor Fuel Rod Behavior, NUREG/CR-0497, TREE-1280, Rev 1, (February 1980).

TABLE A-15.I

UO_2 VISCOSITY DATA FROM TSAI AND OLANDER[A-15.4]

	Temperature (K)	Viscosity (Pa s)
Sample 1	3153	0.00583
	3153	0.00739
	3153	0.00594
	3223	0.00514
	3113	0.00628
	3113	0.00686
	3173	0.00762
Sample 2	3083	0.00921
	3188	0.00869
	3188	0.00771
	3138	0.00781
	3328	0.00602
	3328	0.00765
	3248	0.00808
	3248	0.00682

TABLE A-15.II

UO_2 VISCOSITY DATA FROM WOODLEY[A-15.5]

Temperature (K)	Viscosity (Pa s)
3143	0.00425
3148	0.00365
3153	0.00326
3193	0.00441
3193	0.00434
3193	0.00444
3258	0.00420
3258	0.00417
3258	0.00415
3213	0.00426
3213	0.00428
3218	0.00427
3178	0.00432
3183	0.00436
3183	0.00434
3163	0 00424
3163	0.00420
3163	0.00423
3158	0.00418
3158	0.00428
3163	0.00425
3198	0.00417
3208	0.00418
3198	0.00419
3263	0.00399
3263	0.00405
3263	0.00402
3298	0.00398
3298	0.00395
3303	0.00394
3273	0.00399
3273	0.00398
3273	0.00397
3218	0.00409
3213	0.00406
3218	0.00404
3178	0.00412
3178	0.00406
3178	0.00413

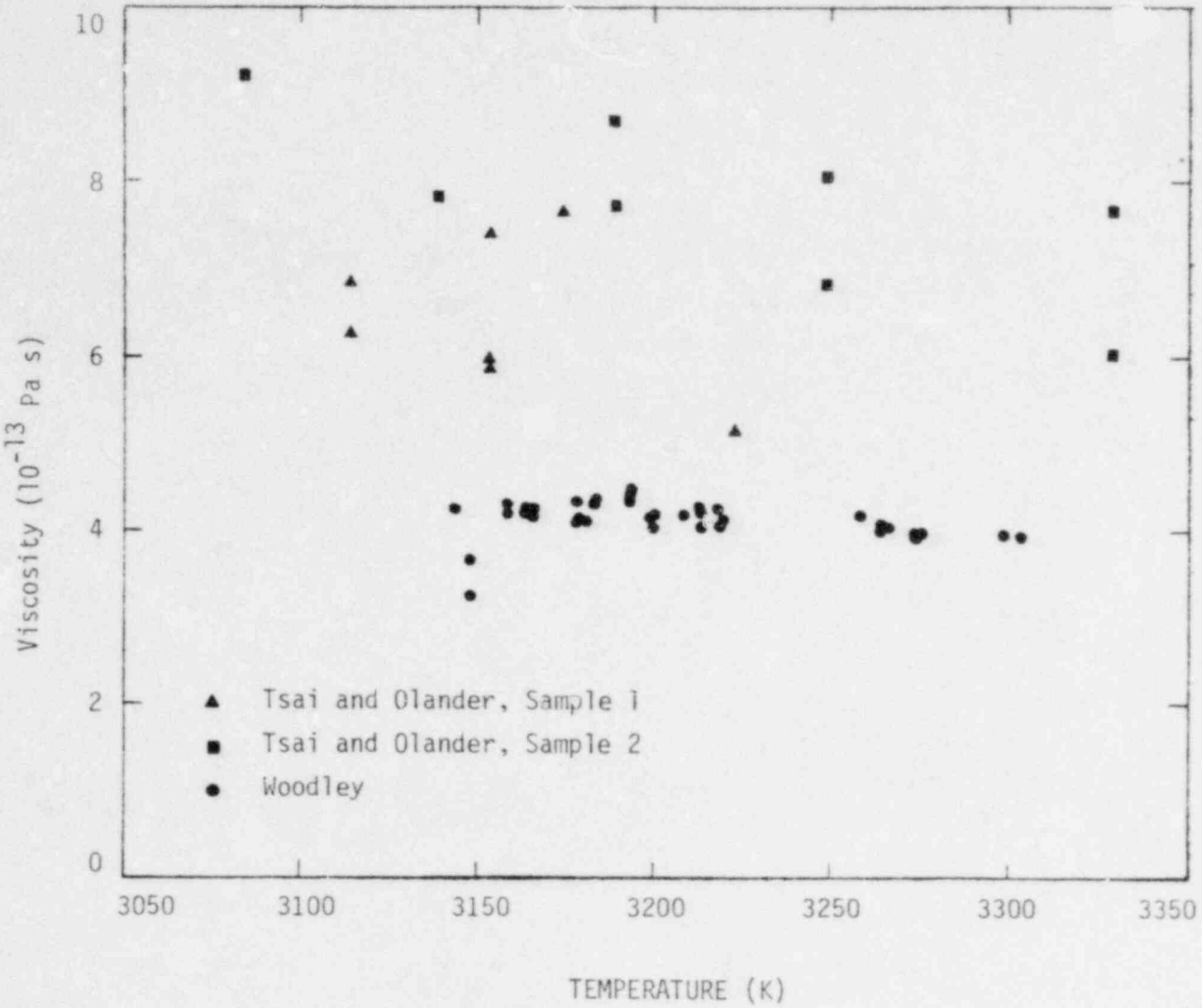

Fig. A-15.1 Uranium dioxide viscosities measured as a function of temperature.

experiments than can be explained by random measurement error. This difference is discussed by Woodley but no definite reason for it was found. The model developed in the next section therefore contains the assumption that the difference between the data of Tsai and Olander and the data of Woodley is caused by some material parameter which has not been considered (oxygen-to-metal ratio, for instance).

15.3 Model Development and Uncertainty

The correlation for the viscosity of UO_2 below the melt temperature was obtained by solving Equation (A-15.5) for the values of the two material constants that reproduce the viscosity measured by Scott, Hall and Williams at 1273 K and the minimum viscosity reported by these authors for uranium dioxide at 1273 K. The fact that this procedure produces only a crude engineering estimate of the viscosity is expressed by assigning a large fractional uncertainty, two thirds, to the predicted viscosity of solid uranium dioxide.

Equation (A-15.1), the correlation for the viscosity of liquid uranium dioxide, was obtained from the data of Tsai and Olander and the data of Woodley. The less precise data of Tsai and Olander were used because Woodley used only one sample and the viscosities measured by Tsai and Olander with their samples differ from Woodley's data by more than the scatter of their measurements.

The traditional Arrhenius relation (Equation A-15.5) was not used to correlate the liquid viscosities because a simpler linear expression fits the data as well as the exponential form. A linear least squares fit to the data of Woodley (with the two anomolously low viscosities at 3148 K omitted) produced the equation

$$\eta_\ell = 1.09\times10^{-2} - 2.09\times10^{-6}\,T \qquad \text{(A-15.6)}$$

The data of Tsai and Olander yielded the following correlation

$$\eta_\ell = 1.60\times10^{-2} - 2.77\times10^{-6}\,T \qquad \text{(A-15.7)}$$

The viscosities predicted by Equation(A-15.6) and (A-15.7) are compared with the data in Figure A-15.2. By inspection of this figure, it was concluded that the best mathematical description of the difference in the viscosities measured for the different lots of uranium dioxide is to assume that the viscosities of the two different lots differ by an additive constant[a].

In order to recognize the more precise measurements of Woodley, yet account for the probable lot-to-lot variation indicated by the data of both authors, the least squares fit to the data of Tsai and Olander was repeated with the added constraint that the slope of the correlation match the slope obtained from the data of Woodley. The resultant correlation for the data of Tsai and Olander is

$$\eta_\ell = 1.38\times10^{-2} - 2.09\times10^{-6}\ T \qquad \text{(A-15.8)}$$

The final step in the derivation of Equation (A-15.1) was to average Equations (A-15.8) and (A-15.6). This step assumes that each lot of UO_2 is equally probable.

The estimated uncertainty of the values of viscosity computed with Equation (A-15.8) was determined using the assumption that the important difference in the measurements of the two references is the unknown difference in the two lots of uranium dioxide. The resultant standard deviation is

$$\sigma = \sqrt{(1.09\times10^{-2} - 1.23\times10^{-2})^2 + (1.38\times10^{-2} - 1.23\times10^{-2})^2}$$

$$\simeq 2\times10^{-3}\ \text{Pa s} \qquad \text{(A-15.9)}$$

which is approximately one third the predicted value of the viscosity.

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[a] The interpretation corresponds to the assumption mentioned at the end of Section 15.2 that the difference in viscosities is caused by some unknown material parameter of the uranium dioxide.

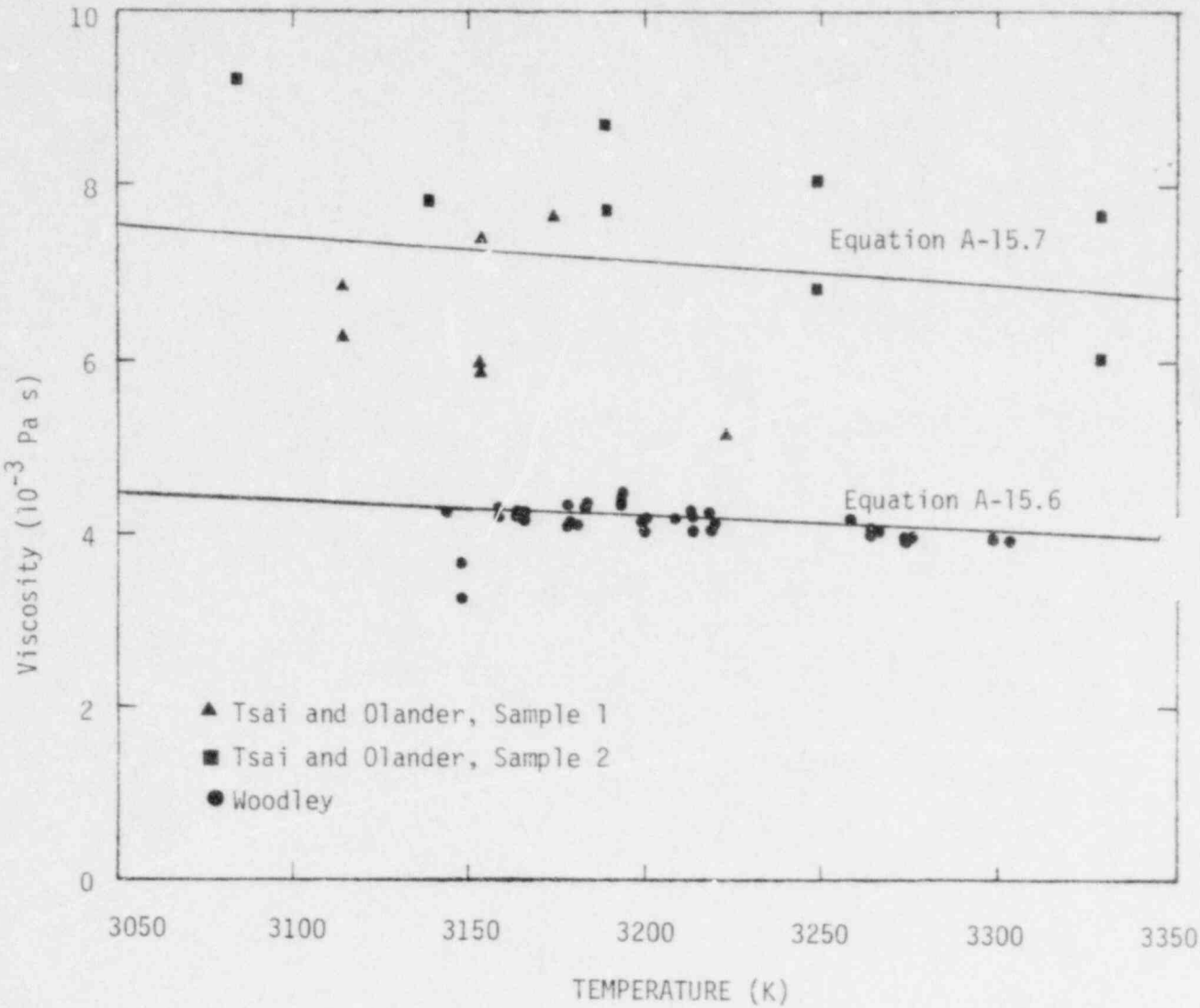

Fig. A-15.2 Data from uranium dioxide samples compared with least squares fit.

The increased uncertainty for non-stoichiometric uranium dioxides shown in Equation (A-15.4) is simply an estimate which has been included to indicate that the model contains no dependence on the oxygen-to-metal ratio of the fuel.

Figure A-15.3 illustrates the viscosities calculated with Equation (A-15.1) for liquid uranium dioxide. The dashed lines are the upper and lower uncertainty limits obtained by adding $\pm$ one third of the predicted viscosity and a melt temperature of 3113 K has been assumed.

Equation (A-15.3), which is employed only in the temperature range where liquid and solid can both exist (that is for temperatures between the fuel melting temperature and the melt temperature plus the liquid-solid coexistence temperature range), is obtained from the assumption that the viscosity is the volume-weighted average of the solid and liquid viscosities in this temperature range.

15.4 Fuel Viscosity Subcode FVISCO Listing

Table A-15.III is a listing of the FVISCO subcode.

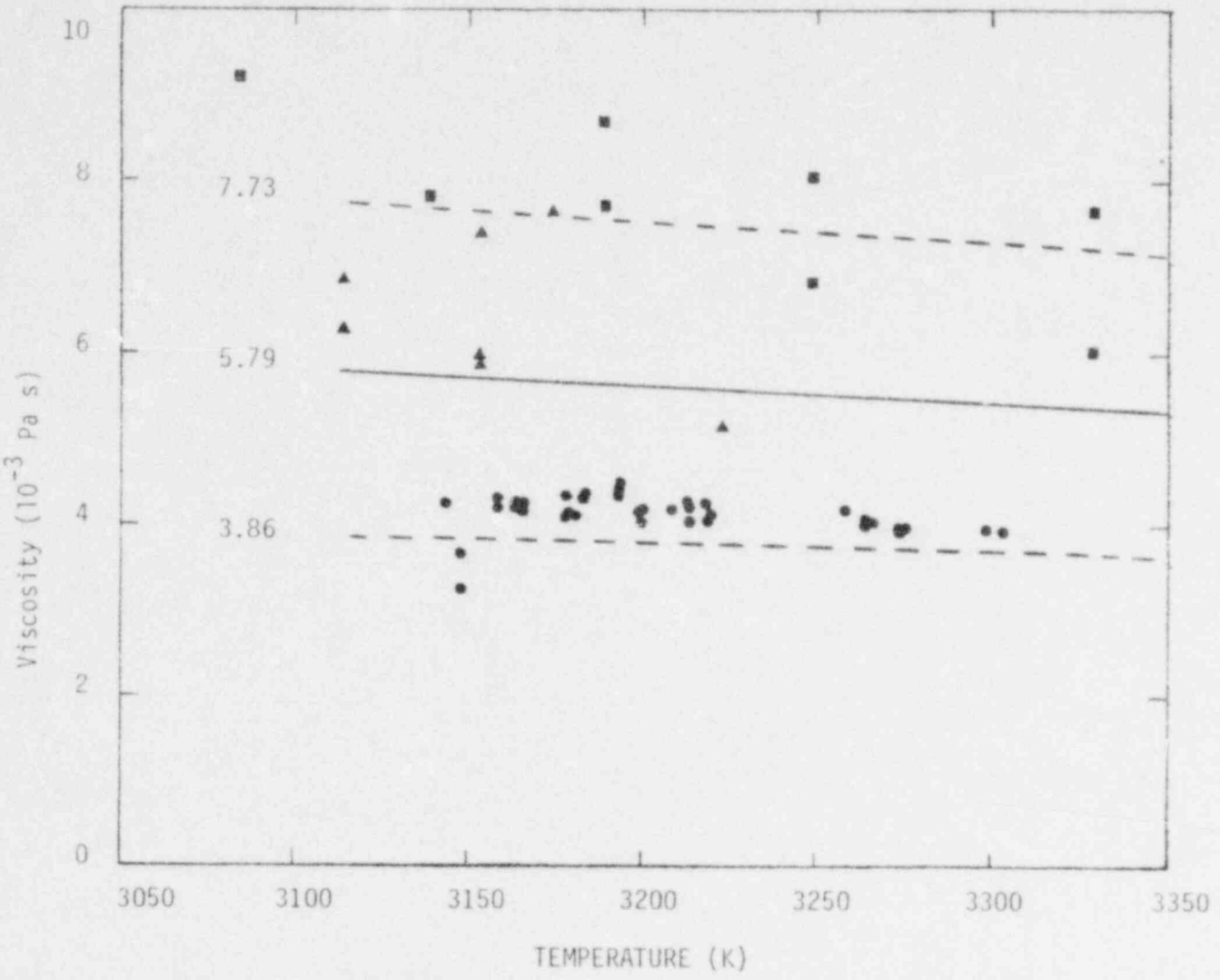

Fig. A-15.3 Viscosities calculated with Equation A-15.1 (solid line) and upper and lower uncertainty estimates (dashed lines) compared with data.

TABLE A-15.III

LISTING OF THE FVISCO SUBCODE

```
      FUNCTION FVISCO(FTEMP,FOTMTL,FACMOT)
C
C     FVISCO CALCULATES THE DYNAMIC VISCOSITY OF UO2 FUEL.
C
C     FVISCO = OUTPUT DYNAMIC VISCOSITY (PA*S)
C
C     UVISCO = ESTIMATED STANDARDD ERROR OF FVISCO (PA*S)
C              (NOT CURRENTLY RETURNED)
C
C     FTEMP  = INPUT FUEL TEMPERATURE (K)
C     FOTMTL = INPUT OXYGEN TO METAL RATIO (UNITLESS)
C     FACMOT = INPUT FUEL FRACTION MOLTEN (UNITLESS)
C              FACMOT = 1.0  MEANS FUEL IS ALL MOLTEN
C              FACMOT = 0.0  MEANS FUEL IS ALL SOLID
C
      COMMON /PHYPRO/ FTMELT   ,FHEFUS      ,CTMELT    ,CHEFUS      ,CTRANB
     *               ,CTRANS   ,CTRANZ      ,FDELTA    ,BU          ,COMP
     *               ,DELOXY
C
C     THE PHYPRO COMMON BLOCK IS SET BY A CALL TO THE SUBROUTINE PHYPRP
C     WHICH IS PART OF THE MATERIALS PROPERTIES PACKAGE USED
C     AT INEL.  QUANTITIES CONTAINED IN PHYPRO USED IN THIS
C     SUBROUTINE ARE:
C
C     FTMELT = FUEL MELTING TEMPERATURE (K)
C     FDELTA = FUEL LIQUID-SOLID COEXISTENCE TEMPERATURE RANGE (K)
C
C     THE EQUATIONS USED IN THIS FUNCTION ARE BASED ON DATA FROM:
C     (1)  R. E. BOOTLEY, THE VISCOSITY OF MOLTEN URANIUM DIOXIDE,
C          JOURNAL OF NUCLEAR MATERIALS,50,(1974),PP 103-106.
C     (2)  F. SCOTT, A. R. HALL, AND J. WILLIAMS, THE PLASTIC
C          DEFORMATION OF URANIUM OXIDES ABOVE 800 C, JOURNAL OF
C          NUCLEAR MATERIALS,1,(1959),PP 39-46.
C     (3)  R. C. TEAL AND J. R. BLANDER, THE VISCOSITY OF MOLTEN
C          URANIUM DIOXIDE,JOURNAL OF NUCLEAR MATERIALS,44,(1972),
C          PP 93-96.
C
      FVISCO = 2.38 * EXP(9.542E+04/FTEMP)
      IF (FTEMP .LE. FTMELT) GO TO 120
      FVISCO = FVISCO * (1. - FACMOT) + (1.23E-02 - 2.09E-06 * FTEMP)
     *         * FACMOT
      IF (FTEMP .GE. (FTMELT + FDELTA)) GO TO 100
      FVISCO = 1.23E-02 - 2.09E-06 * FTEMP
  100 UVISCO = FVISCO*0.33 * (1. + ABS(FOTMTL - 2.0))
  120 IF (FTEMP .LT. FTMELT) UVISCO = FVISCO*0.67 * (1. + ABS(FOTMTL -
     *    2.0))
      RETURN
      END
```

15.5 References

A-15.1 R. Scott, A. R. Hall and J. Williams, The Plastic Deformation of Uranium Oxides above 800°K, Journal of Nuclear Materials 1, pp 39-48 (1959).

A-15.2 W. F. Sheely (Ed)., Quarterly Progress Report July, August, September, 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1223 (November 1969).

A-15.3 W. F. Sheely (Ed)., Quarterly Progress Report, October, November December, 1969, Reactor Fuels and Materials Development Programs for Fuels and Materials Branch of USAEC Division of Reactor Development and Technology, BNWL-1279 (February 1970).

A-15.4 H. C. Tsai and D. R. Olander, The Viscosity of Molten Uranium Dioxide, Journal of Nuclear Materials, 44 pp 83-86 (1972).

A-15.5 R. E. Woodley, The Viscosity of Molten Uranium Dioxide, Journal of Nuclear Materials, 50, pp 103-106 (1974).