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The viscosity of UO_2 is needed to analyze gross fuel movement in various potential accident situations in LMFBR.[1] This transport property has been measured from the melting point to 3000°C using an oscillating cup viscometer. The apparatus and method of data analysis have been described previously.[2] A cylindrical crucible containing the sample is suspended in a vacuum furnace by a wire. The assembly is given an initial twist to displace it from the equilibrium position. The resulting motion is one of damped oscillation of a torsional pendulum. The period and decay constant of the oscillation are directly related to the liquid viscosity, which supplies the viscous drag that causes damping of the oscillation.

Approximately 80 grams of UO_2 was contained in a 0.31 cm wall thickness, 1.6 cm i.d. tungsten crucible bored from forged rod stock. The lid and support rod were electron-beam welded onto the crucible under vacuum. Prior to sealing, the UO_2 was baked in a hydrogen atmosphere at 1600°C for ten hours, which should produce stoichiometric or slightly hypostoichiometric material.[3]

The major modification of the technique described in Ref. 2 was the method used to measure the parameters of the damped sinusoidal oscillation. A laser beam reflected from a polished surface on the oscillating crucible assembly periodically triggered a photocell in the vacuum tank. The output pulses from the photocell generated by passage of the reflected laser beam advanced the channel of a multichannel analyzer operating in the multiscalar mode. At the

same time, 100kHz timing pulses were fed into the multichannel analyzer. The number of such timing pulses stored in a particular channel provided an accurate measure of the time elapsed between successive passages of the laser beam past the photocell. Fig. 1 shows a typical set of data obtained in this manner. The position of the photocell was different from the position of the reflected light beam with the pendulum at equilibrium. The upper points represent the time interval for the pendulum to move from the photocell to the maximum angular displacement farthest from the photocell and return to the photocell. The lower points are a similar record for the remaining portion of each oscillation (i.e., from the photocell to the maximum displacement nearest the photocell and back). A total of 27 oscillations were recorded before the amplitude of the oscillation was insufficient for the reflected light beam to reach the photocell. By analysis of the variation of these time intervals for a number of oscillations, the frequency and logarithmic decrement of the damped oscillations were obtained. The viscosity was obtained from these data by the usual method.[2]

The entire system was operated in vacuum in order to prevent attack of the crucible and the furnace element by gaseous impurities, to eliminate any viscous drag on the oscillating system due to the surrounding gases and to reduce the heat loss by convection. A mechanical pump and two sorption pumps were sufficient to handle the outgassing from the system at high temperature. To avoid vibration, only the sorption pumps were left on when taking data. Pressures of $\sim 2 \times 10^{-4}$ torr were achieved in this manner.

Temperatures were measured by an optical pyrometer calibrated

against an NBS secondary standard up to 2500°C. The calibration was extended to ∿ 3000°C by measuring the temperature at which a tantalum wire passing through the hot zone of the furnace melted. The pyrometer reading, after correction for the window and prism in the light path, agreed to within 15°C of the known melting point of tantalum. During operation, a movable flag prevented coating of the prism by tungsten vapor from the furnace.

Measurements were made on two different samples of UO_2 with the results shown in Table 1. Sample #1 was tested twice with solidification and remelting in between the two experiments. The intrinsic decay constants (i.e., system damping with the sample solid) ranged from 10% to 25% of the total decay constant and were determined for each run. The solution of the momentum equations for the oscillating cylinder permits two viscosity values for each measured decay constant. The smaller values were chosen because the measured decay constants decreased with temperature, which is consistent with a positive activation energy for viscosity.

A least squares fit of the data plotted in Fig. 2 yields a viscosity at the melting point (2800°C) of 8 ± 1 cp. The average deviation of the data from the best fit straight line on Fig. 2 is ± 1 cp. The viscosity measured here is considerably less than either the value of 25 cp currently assumed for accident analysis[1] or the 36 - 46 cp measured by Bates et al.[4]

The activation energy is 15 ± 8 kcal/mole. This poor precision is due to the scatter of the data and the rather small temperature range over which measurements were made.

The meniscus effect[4] of the liquid UO_2 in the crucible, which

tends to increase the apparent viscosity, was neglected in the calculation. One of the crucibles was sectioned longitudinally after the experiments. Visual examination of the solidified UO_2 indicated that the meniscus increased the height of the contact between the UO_2 and the crucible wall by about 5%. This figure represents the maximum error in the viscosity due to curvature of the upper surface of the melt.

As can be seen from Table 1, the UO_2 appears to have been liquid at a temperature as low as 2780°C, which is 85°C lower than the melting point of stoichiometric urania recently reported by Latta and Fryxell[6]. This discrepancy may have been due to: (1) temperature measurement errors due, for example, to coating of the pyrometer prism by tungsten; (2) reduction of the stoichiometry by the hydrogen bake; or (3) contamination of the melt by the crucible material. Possibilities (2) and (3) cannot both occur, since tungsten solubility in molten urania decreases rapidly as the O/U ratio is reduced[6]. Visual examination of the sectioned crucible showed no signs of interaction between the melt and the crucible. In regions where the UO_2 had separated from the wall, the tungsten was very smooth and shiny.

Acknowledgments

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Table 1

UO_2 VISCOSITY DATA

Sample #1

Mass: 93.6 gms.

Moment of Inertia: 823.0 gm-cm^2

Torsion Wire: 8 mil molybdenum

Radius: 0.79 cm

Temperature (°C)	Decay const. ($\times 10^3$ sec^{-1})	Period (sec)	Density* (gm/c.c.)	Viscosity (cP)
First Melt				
2880	4.67	5.14	8.71	5.83
2880	5.10	5.12	8.71	7.39
2880	4.70	5.12	8.71	5.94
2940	4.47	5.13	8.66	5.14
Second Melt				
2825	4.73	5.23	8.77	6.28
2825	4.90	5.21	8.77	6.86
2880	5.10	5.21	8.71	7.62

Sample #2

Mass: 72.0 gms.

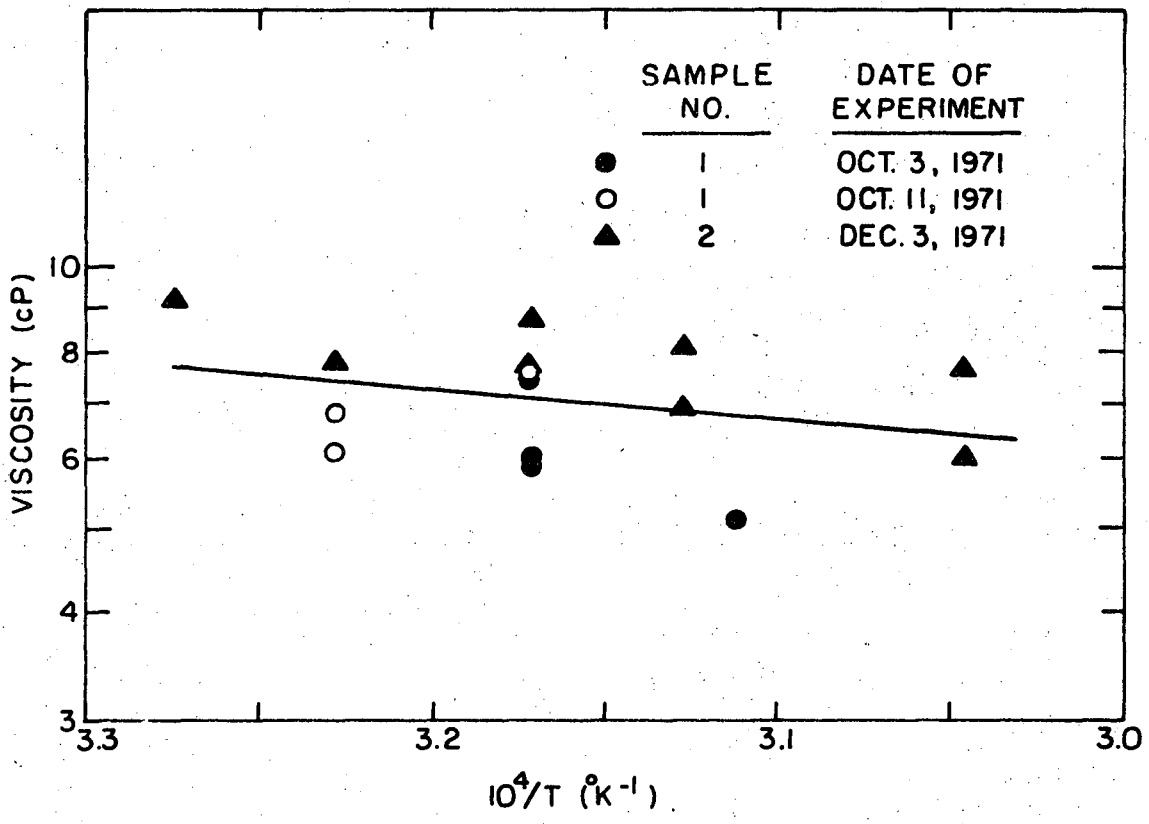
Moment of Inertia: 547.9 gm-cm^2

Torsion Wire: 8 mil molybdenum

Radius: 0.81 cm

Temperature (°C)	Decay const. ($\times 10^3$ sec^{-1})	Period (sec)	Density* (gm/c.c.)	Viscosity (cP)
2780	7.39	4.26	8.80	9.21
2880	7.32	4.24	8.71	8.69
2880	7.00	4.24	8.71	7.71
2825	7.01	4.26	8.77	7.81
3010	6.41	4.26	8.59	6.02
3010	7.00	4.26	8.59	7.65
2925	7.14	4.24	8.67	8.08
2925	6.70	4.24	8.67	6.82

*taken from Refs. 4 and 5



XBL 721-5913

Fig. 1

A typical picture of stored counts vs. channel number. Each vertical scale unit represents 100 K counts (1 sec.).

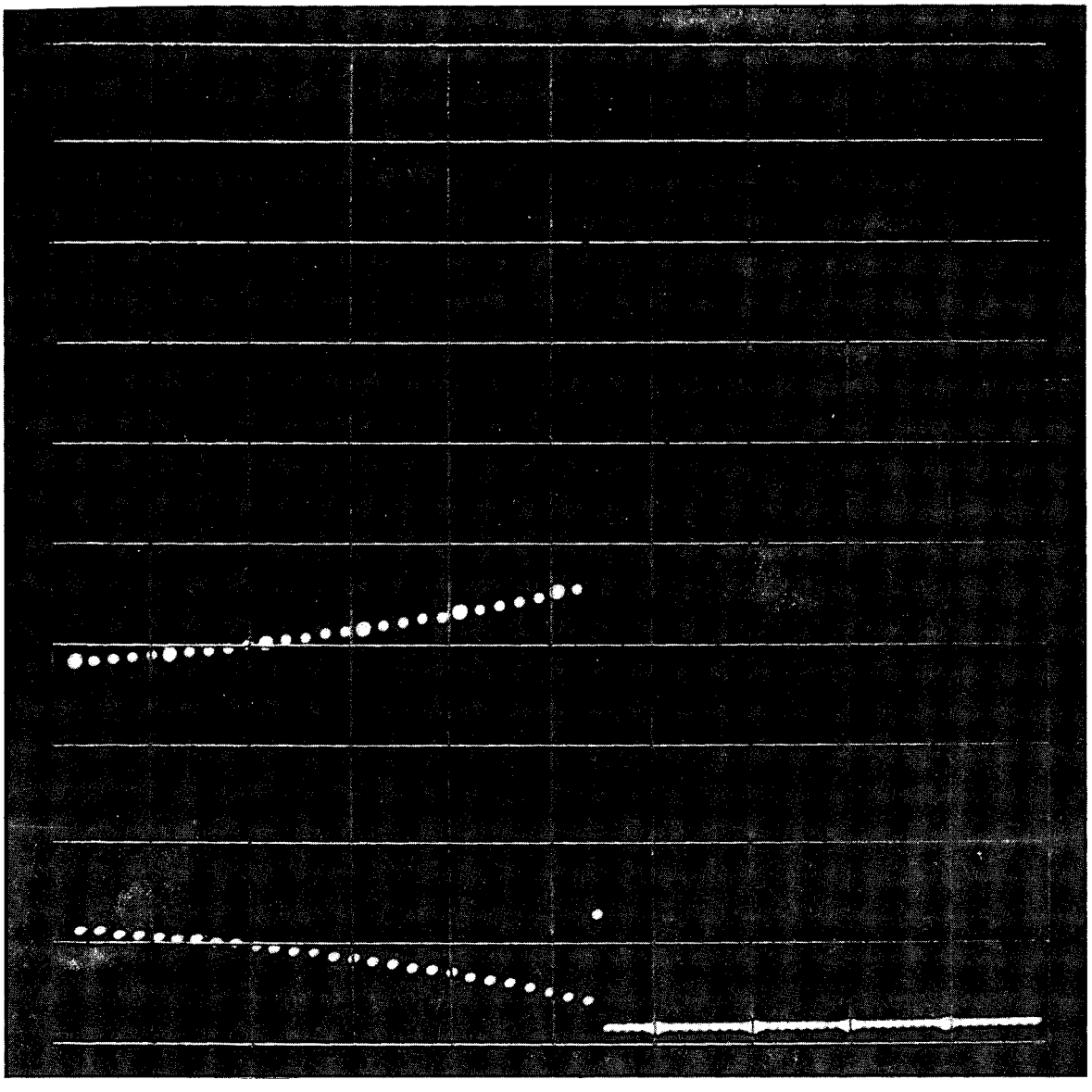


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Fig. 2

Viscosity of liquid UO_2 vs. temperature.

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