An Experimental and Analytical Study of Heat Transfer Characteristic of a UF_6-Filled Vessel

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Abstract

Experimental and analytical study was conducted aiming at a better understanding of thermal behavior of uranium hexafluoride in cylinders. The experimental study includes the measurement for thermal conductivity of solid and liquid uranium hexafluoride as well as the thermal test for a bare vessel filled with about 110 kg of uranium hexafluoride.

The thermal conductivity of solid and liquid uranium hexafluoride was measured by steady and non-steady methods and was found to be in fairly good agreement with the values estimated by the Weber's empirical equation. The thermal test was conducted to observe the phase changes of inner uranium hexafluoride as a function of time and location as well as the apparent heat transfer coefficient between the cylinder material and inner uranium hexafluoride.

Based on both the observation of this thermal test and the experimental determination of thermal conductivity, the two-dimensional analysis is carried out to complete liquefraction of uranium hexafluoride in the test cylinder by using a Phoenics code which takes direct account of fluid flow effect.

Introduction

The main purpose of this study is to obtain a better understanding of thermal properties of uranium hexafluoride in cylinders. The study includes thermal test for a test cylinder filled with about 110 kg of uranium hexafluoride as well as the experimental determination of thermal conductivity of solid and liquid uranium hexafluoride by steady and non-steady methods.

The measurement of thermal conductivity may serve as basic data for safety assessment of uranium hexafluoride cylinders under fire, while the thermal test for a test cylinder filled with actual uranium hexafluoride gave us useful information about phase changes of uranium hexafluoride as a function of time and location in a test cylinder. These experimental findings will contribute to determine analytically if the cylinder would hydrostatically rupture and the time available for fire fighting before the incident occurred.

Experimental Determination of Thermal Conductivity of UF_6

Measurement of thermal conductivity of uranium hexafluoride is rarely to be found except for one measured at 72°C by Priest(1), although it is a very important property in solving heat transfer problem of a uranium hexafluoride cylinder. Measurements are under way by both steady and non-steady methods. Only a part of the results is described in this report, because the experiment is not fully done from the difficulty in measuring accurate temperature changes or temperature differences due to highly sublimating nature and large volume change from solid to liquid of this material.

Two separate sets of experimental apparatuses were prepared for the steady and non-steady measurements. Each apparatus was used for both solid and liquid measurements.

Shown in Fig.1 is a plate apparatus for the steady measurement, where uranium hexafluoride was introduced into a gap space between the reference plates. CaF_2 glass and the stainless steel SUS-304 were chosen as the reference materials, of which thermal conductivities were well studied. The upper part was kept at higher temperature by hot water jacket and the lower part was kept at lower temperature by coolant. The apparatus has a window at a lower part which allows us to observe the phase change of uranium hexafluoride inside. The measuring part of this apparatus was encased in a heat insulator to maintain a homogeneous temperature distribution. A hot-wire cell as shown in Fig.2 was to determine thermal conductivity by transitory temperature method. The hot wire was 0.2 mm in diameter and was Chromel A (80%Ni-20%Cr), of which electric resistance remains unchanged with temperature change.

The thermal conductivity is derived from observed temperature differnce in steady method or from observed temperature gradient with time in non-steady method by using the following equations:

For the steady method,

$$\lambda_s = \lambda_r (\Delta T_r / \Delta L_r) / (\Delta T_s / \Delta L_s)$$

where λ is the thermal conductivity, ΔT is the temperature difference observed in the distance ΔL, and the suffixes **r** and **s** refer to the reference material and the sample material, respectively.

For the non-steady method,

$$\lambda = (Q / 4\pi \Delta T) \ln (t_2 / t_1)$$

where **Q** is the heat generated by a unit length of a hot wire in a unit duration and ΔT is the temperature change from time t_1 to t_2.

Experimentally determined values for thermal conductivity of both solid and liquid uranium hexafluoride are plotted in Fig.3. For comparison, thermal conductivity measured by Priest is also shown in the same figure.

Another assessment may be given by the Weber's empirical equation(2),(3), which allows us to derive thermal conductivity from specific heat and density as follows.

$$\lambda = A C_P \rho (\rho / M)^{1/3}$$

where **A** is assumed to be, for any substances, a constant value, which is proposed to be $3.59X10^{-3}$ by Weber (2),(3), and Kuong(4) and to be $4.30X10^{-3}$ by Smith(5). As shown in Fig.3, this empirical equation also implies that the thermal conductivity obtained in the present work lies within a reasonable range of values. Further accumulation of experimental data will be made to clarify the temperature dependence of thermal conductivity by eliminating several minor problems in the present measurement.

Thermal Test of a UF_6-Filled Vessel

Detailed results of the thermal test was described elsewhere(6), and only a brief description will be given here. As shown in Fig.4, the equipment for thermal test consists of a 270 mm in diameter, 1400 mm long and 30 mm thick carbon steel test cylinder encased in a 20 kW electric heater and measuring sensors. The test cylinder has a valve on its end plate to imitate an actual cylinder and was filled with about 110 kg of uranium hexafluoride, which amounted to 95% in volume of the test cylinder when the temperature of inner uranium hexafluoride became 120°C. The heater was encased in the reflector and the insulator to maintain uniform heating. The heating apparatus was controlled by a PID controller within the temperature range from 80° to 400°C on the heater surface.

As for the measuring sensors, there were 28 conventional sheathed thermocouples to measure temperatures of various places of the equipment and a MKS 5034 capacitance manometer to measure inner pressure of the test cylinder. Among 28 thermocouples, two sets of five thermocouples were for temperature measurements of the heater surfaces and the valve. Temperatures of the outer and inner surfaces of the cylinder as well as temperatures of uranium hexafluoride inside the cylinder were measured by the other 18 thermocouples. These 18 thermocouples were divided into several groups to measure the radial temperature distribution. We labeled the groups measuring the lower and the upper parts on the vertical center line as "A"("E") and "D", respectively. Labels "B" and "C" were used for the groups measuring the parts with angles of 90° and 45° to the vertical center line. These labels had additional suffixes to indicate radial positions. The outer and inner surfaces of the test cylinder were indicated by the suffixes "1" and "2", respectively. Indicated by "3", "4", "5" were 3 mm, 23 mm and 43 mm inwards away from the inner surface. As shown in Table 1, the total of 11 runs of thermal test were carried out with the maximum temperature of the heater set for 200, 300 and 400°C.

Shown in Fig.5 is a typical example of the results in the thermal test. We can see that even after the heater surfaces reaches the predetermined temperature, 200°C in this case, the inner pressure goes up rather slowly and then shows a plateau at 1137 torr, which is in good agreement with the triple point vapor pressure of uranium hexafluoride and thus seemed to be the indication of the onset of liquefying. After staying here in some period, the pressure rises rapidly far beyond the triple point, while most of the uranium hexafluoride in the test cylinder is considered to remain still in solid state judging from the temperature of TE-3, which is lower than the triple point temperature of uranium hexafluoride, 64°C. The solid uranium hexafluoride in this region gradually melts a while later. One more to be mentioned is the temperature drop of the inner surface right after the inner pressure reaches the triple point vapor pressure of uranium hexafluoride. This trend is much clearer in the bottom part of the test cylinder, whose temperature shows a sharp decrease to 64°C, as shown in TA and TE. This temperature drop is considered to be caused by liquid uranium hexafluoride flowing down to the bottom. In a 400°C operation, similar trend was observed as shown below in the same figure. We also observed that the temperature difference between the cylinder material and inner substance increased with time when the temperature of inner substance remained lower than its triple point temperature and then observed that this temperature difference disapperared when uranium hexafluoride turned into liquid. This led us to assume a gap conductance between the inner surface of the cylinder and uranium hexafluoride.

Numerical Analysis of Thermal Behavior of a UF_6-Filled Vessel

The hydrostatic rupture concept of the cylinder caused by the expansion of liquid uranium hexafluoride under fire was usually based on the following assumption. The heat entering through the cylinder wall would make the uranium hexafluoride melt in an equilibrium state at the triple point. And it would be not until the whole uranium hexafluoride became liquid that the inner pressure and the temperature started to increase over the triple point ones. However, the result of thermal test indicates the different features as stated in the preceding paragraph.

We have reported(6) that a two-dimensional analysis based on heat transfer model gave a fairly good reproducibility of thermal test, even when we assumed the heat transfer values derived from a one-dimensional analysis in three directions, which is shown in Tables 2 and 3. However, the reproducibility of this method became poor at higher temperature and furthermore it was found to need a long CPU time to proceed the calculation to complete liquefaction. To eliminate the latter problem, we carried out a two-dimensional analysis by using a Phoenics code which takes direct account of fluid flow effect assuming the following model. We take account of the flow effect and volume expansion effect of liquid uranium hexafluoride and also take account of the gap conductance observed between the inner surface of the cylinder and solid uranium hexafluoride, which will disappear by the onset of liquefaction. However, the overall emissivity on the inner surface and the latent heat of vaporization are neglected because of their small contribution. One more to be mentioned is that we take the thermal conductivity of liquid uranium hexafluoride derived from Weber's empirical equation, which is found to be in good agreement with our experimental determination of this property.

Fig.6 shows a thermal model and a mesh configuration used in the present calculation by using a Phoenics code. This configuration represents only a right half of the test cylinder cross section based on a symmetric assumption. The mesh division is intended to be suitable for heat flux calculation and for taking into account of natural convection effect.

The calculated temperature change with time is shown in Fig.7. For comparison experimentally observed temperature is also plotted in the same figure. A roughly good agreement is obtained between the calculation and experiment, but the agreement is poor for TB's, which represent the temperatures right below the upper surface of uranium hexafluoride. This disagreement may be due to that experimentally we observed the temperature of liquid uranium hexafluoride, while in the calculation the uranium hexafluoride remains still solid because of negligence of the gravitation of solid uranium hexafluoride in its liquid. Another reason for the disagreement may be due to that we take a simple two-phase model for uranium hexafluoride inside the cylinder which forms actually a complicated solid, liquid and vapor phases. Modification must be made in computing the thermal behavior of uranium hexafluoride inside the cylinders, while in the present method the computation can be carried out to complete liquefaction of uranium hexafluoride within a reasonable CPU time.

References

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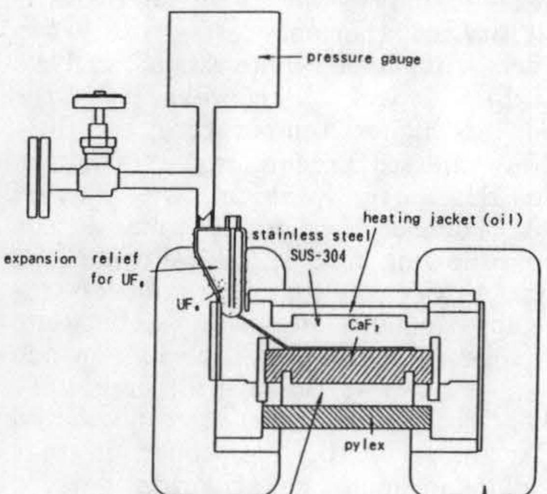

Fig.1 Plate apparatus for steady measurment CaF_2 glass and stainless steel SUS-304 are the reference materials.

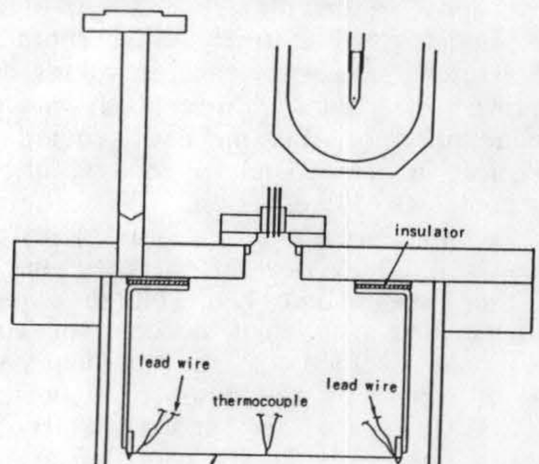

Fig.2 Hot-wire cell for non-steady measurement Hot-wire is 0.2mm in diameter and is Chromel A, whose electric resistance remains unchanged with temperature change.

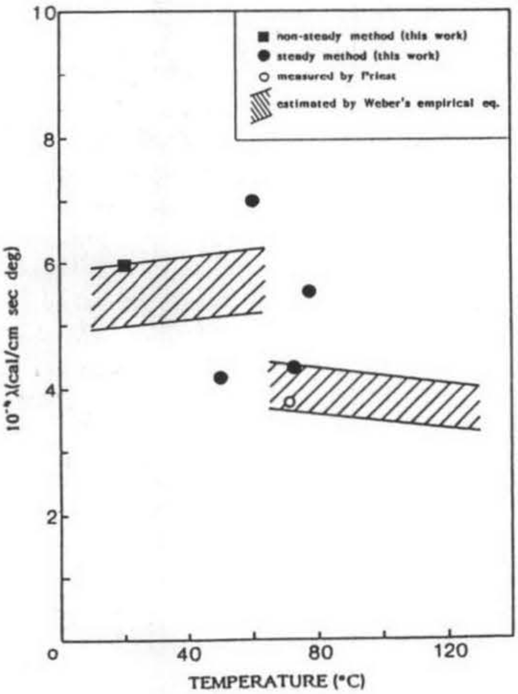

Fig. 3 Experimentally determined thermal conductivity of uranium hexafluoride. Black circles and square were measured by steady and non-steady, respectively.

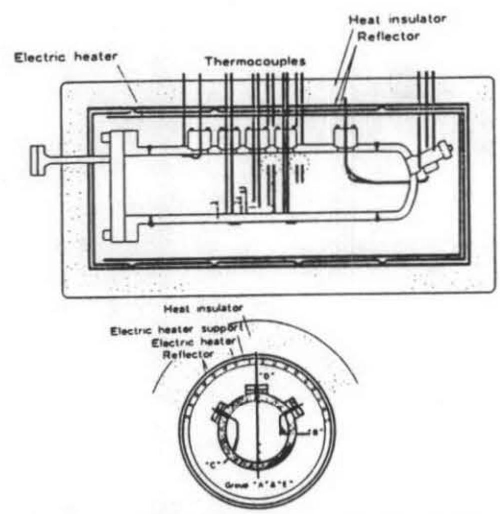

Fig. 4 The test cylinder encased with electric heater, reflector and heat insulator. The figure also shows thermocouples for temperature measurement which are divided into several groups indicated by "A", "B", "C", "D" and "E".

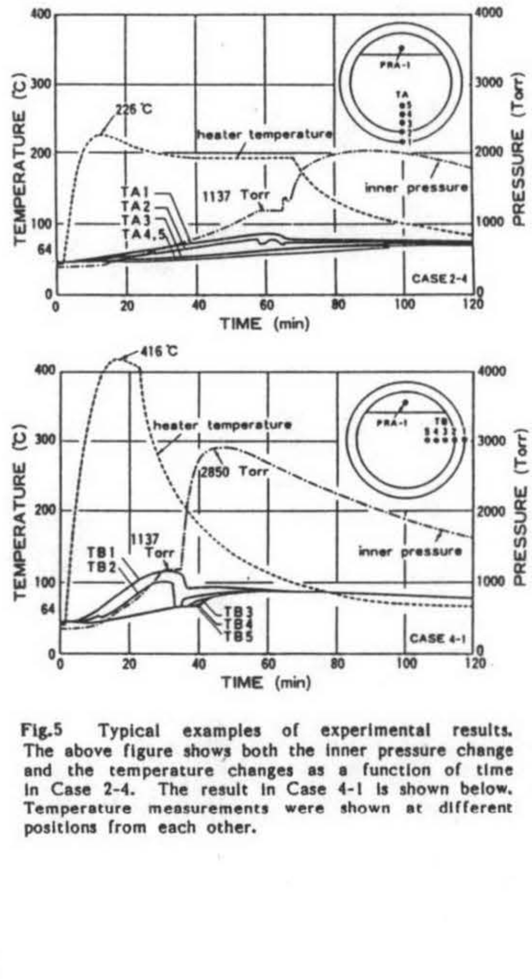

Fig.5 Typical examples of experimental results. The above figure shows both the inner pressure change and the temperature changes as a function of time in Case 2-4. The result in Case 4-1 is shown below. Temperature measurements were shown at different positions from each other.

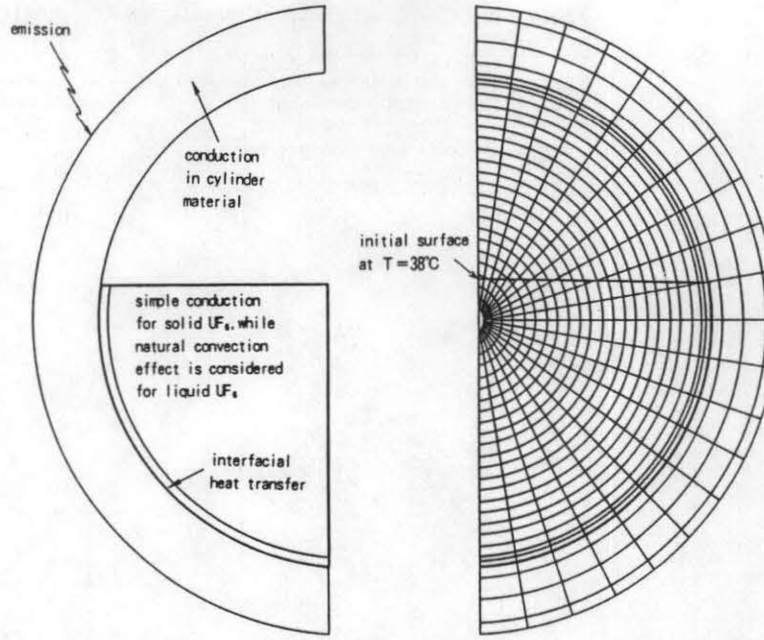

Fig.6 Heat transfer model and mesh division used in two-dimensional analysis by Phoencs.

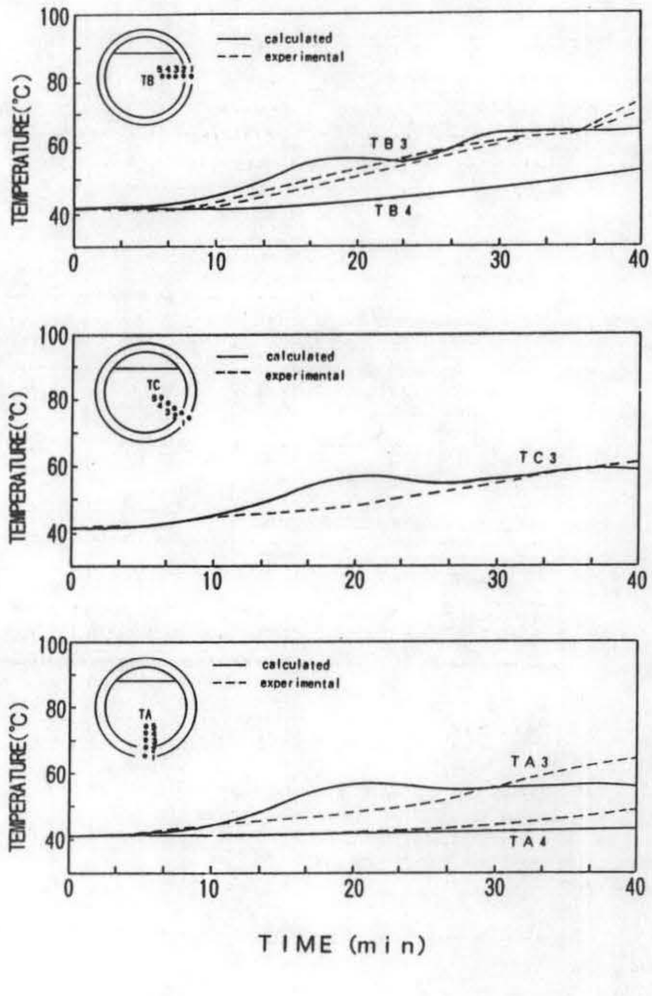

Fig.7 Time dependence of inner uranium hexafluoride temperature simulated for the thermal test of T=400°C on the heat surface by two-dimensional analysis.

Table 1. Conditions of thermal tests carried out in August 1987.

Case No.	UF_6 condition		Thermal condition		
	weight (kg)	initial form	heater temp. (℃)	heating rate (℃/min)	heating time (min.)
1	112		400	36	43
2 - 1	107		200	27	42
2 - 2					56
2 - 3					60
2 - 4					60
3 - 1	107		300	36	26
3 - 2					21
3 - 3					20
4 - 1	107		400	36	10
4 - 2					17
4 - 3					25

Table 2. Overall emissivity on the cylinder surface and heat transfer coefficient between inner surface of the cylinder and uranium hexafluoride , derived through one dimensional analysis using the experimental data.

Direction (θ)	heat transfer coefficient (kcal/m² h ℃)								overall emissivity (−)
	temperature (℃)								
	0	53	60	62	64	70	72	85	
A	11	11	19	27	74	—	—	—	0.55
B	15	15	23	25	30	210	330	1150	0.60
C	10	10	17	22	52	—	—	—	0.58

Table 3. Apparent heat conductivity of uranium hexafluoride obtained by one dimensional analysis.

Description	apparent heat conductivity of UF_6 (k cal/m h ℃)							
	temperature (℃)							
	35	45	55	59	63	64	65	69
This work	0.05	0.73	0.97	1.23	1.80	2.25	—	250
PATRAM '83	—	—	2.58	—	—	—	3.44	—