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UO_2の降状応力のひずみ速度および温度依存性

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Strain Rate and Temperature Dependence of Yield Stress of Uranium Dioxide

Toshimichi TACHIBANA*
Hirotaka FURUYA*
Masumichi KOIZUMI*

Abstract

Specimens of polycrystalline UO_2 have been strained in compression at constant strain rate from 0.05 to 10.0 /min and at temperatures from 900° to 2,000°C. Results are presented showing the characteristic yield points and the variation of the yield stress with strain rate and temperature. The experimental data shows that the values of strain rate sensitivity and activation energy for yield flow of UO_2 at high strain rate are greater than that of previous data which was given at usual strain rates.

The data are discussed in relation to the mechanism which controls the yield strength of UO_2. The yield strength at high strain rate may be controlled primarily by the Peierls stress.

* Research and Development Section, Plutonium Fuel Division, Tokai Works

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Strain Rate and Temperature Dependence of Yield Stress of Uranium Dioxide.

実施責任者 小 泉 益 通*
報 告 者 立 花 利 道*
古 屋 廣 高*

要 旨

多結晶UO_2ペレットを900°から2,000℃の温度範囲で，0.5から10.0／min の広範囲のひずみ速度で圧縮変形した。この結果，著しい降状現象が観測された。さらに，UO_2の降伏強さの温度およびひずみ速度依存性がえられた。この実験結果は，UO_2が降状変形するときの活性化エネルギーとひずみ速度敏感性の値が，従来通常のひずみ速度で測定されてきた値よりも高かった。この測定結果をもとに，UO_2の降状強さを主として規制する機構に関して考察した結果，高ひずみ速度のもとでのUO_2の降状強さは，おもにパイエルス力によって制御されるだろうという結論がえられた。

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I. INTRODUCTION

The recent development of analytical models of fuel element performance has led to an awareness of the need for mechanical property data on nuclear fuel, particularly yield point because deformation behaviours of nuclear fuel suddenly change at yield point(1). The yield characteristics of sintered UO_2 have been investigated as function of temperature, strain rate and stoichiometry by several workers(2)~(6).

The first measurements of the yield strength were made by Byron(2) and Nadeau(3) by using the compression method at temperatures from 600° to 2,000°C and 900° to 2,100°C, respectively. Thereafter, Evans et al.(4) and Canon et al.(5) measured the elastic limit by using the bending method at temperatures from 800° to 1,400°C and 1,000° to 1,800°C, respectively. Moreover, Roberts et al.(6) measured the influence of porosity on the elastic limit at 1,250° and 1,600°C by the bending method. Recently, Guerin(7) measured the yield stress by using the compression method at temperatures from 600° to 1,700°C. The data obtained by the above mentioned workers agree with each other in the experimental fact that the yield stress of UO_2 strongly depends on the temperature and the strain rate. However, they have different opinions about the mechanism which would primarily control the yield stress of UO_2.

The purpose of the present study is to provide additional information which might better identify the primarily controlling mechanism of the yield strength of UO_2. Polycrystalline specimens of nearly stoichiometric UO_2 were deformed in compression in the wide strain rate range (0.05 to 10.0 /min) and at temperatures from 900° to 2,000°C. The strain rates in the present study were higher than those of the previous workers(2)~(9).

II. EXPERIMENTAL

1. Test specimens

UO_2 pellets, approximately 5.5 mm in diameter and 10 mm in height, were obtained from Nuclear Fuel Ind. Ltd. They were fabricated by cold-pressing the ball-milled UO_2 powder and subsequently sintering the compacts in a reducing atmosphere (75%H_2-25%H_2). The average density, grain size and O/U ratio of the specimens are 94±1% theoretical, 17μ and 2.00±0.01, respectively. The chemical analysis of one the specimens is given in Table 1. The surfaces of some specimens were polished on fine emery papers (SiC#400) and lapped on a revolving cloth-covered wheel with finer abrasive (Al_2O_3), and those polished surfaces were etched with a chemical solution before compression test.

2. Testing procedure

The schematic diagram of the testing instrument was shown in Fig. 1. The compression tests of the specimens were carried out in a vacuum of about 10^{-5} torr at the temperature range from 900° to 2,000°C by using an Instron type testing machine having crosshead speeds suitable for strain rates in the range from 0.05 to 10.0 /min. The heating furnace comprises tantalum band heaters with some tantalum band reflectors. The temperature of the furnce was increased at a constant rate of about 50°C/min to the desired value, and then maintained for 15 min before starting the compression test. The temperature of the specimen was measured by a micro-pyrometer. Below 1,500°C, an additional measurement was made by touching a thermocouple (Pt/20%Rh-Pt/40%Rh) to the circumferential side of the specimens. The optical pyrometer was calibrated at the melting points of Ag, Au, Cu, Fe, Pt and Al_2O_3. The strain rate of 0.1/min was used for the standard test of all specimens and strain rates of 0.05, 0.1, 1.0 and 10.0/min for the measurement of the strain rate dependence on the yield stress. The specimens which had been polished and etched were examined by means of an electron microscope to see the exsistence of slip lines after the compression test. The electron micrographs were made on a Hitachi HS-7 microscope at an accelerating voltage of 50 Kv by means of a two-stage replica technique.

III. RESULTS

Figure 2 shows the stress-strain curves of UO_2 obtained at a strain rate of 0.1/min in the temperature range from 900° to 2,000°C. Two types of curves can be seen. Below 1,400°C, the characteristic yieldings are observed, indicating the distinct lower and upper yield points as typically shown in Fig. 3. Above 1,600°C, on the other hand, the stress increases with strain and the two yield points are indistinguishable.

The curves in Fig. 4 show the temperature dependence of the yield stress for UO_2, where the yield stresses are defined by the lower yield stress (σ_{ly}) in the case of the characteristic yielding and by the stress (σ_y) shown in Fig. 5(b) in the other case. It can be seen that the yield stress decreases with increasing the temperature and particulary above

1,900°C decreases abruptly.

The additional measurements of the yield stress were carried out in the wide range of the strain rate. Figure 6 shows the variation of the yield stress with the logarithm of the strain rate ($\dot{\varepsilon}$). The yield stress strongly depends on the strain rate.

When the deformation is assumed to be controlled by a single thermally-activated process, the effective activation energies (H) can be determined from the results shown in Fig. 4 and 6, using the following equation(2) (3) (20),

$$H = -kT^2(\partial\sigma/\partial T)_{\dot{\varepsilon}}\cdot(\partial ln\dot{\varepsilon}/\partial\sigma)_T \quad (1)$$

where k and T are Boltzmann's constant and absolute temperature, respectively. Table 2 shows the results obtained by the above mentioned procedure. It can be seen that the activation energy is independent of the temperature and does not agree with the Byron's results.

IV. DISCUSSION

There have been three models to explain the mechanism which primarily controls the strength of UO_2. The first is the diffusion mechanism, in which the yield strength is controlled by vacancy diffusion to grain-boundary and its subsequent sliding.

Canon et al.(5) measured the high temperature deformation of UO_2 by using the four point bending method, and showed that the activation energies obtained at proportional limit agree with the acivation energy for uranium self-diffusion(12) for UO_2 in the plastic deformation region and that the strength was associated with the onset of grain-boundary sliding.

Roberts et al.(6) also measured the activation energies at 1,250° to 1,600°C, which showed good agreement with the energy for uranium self-diffusion in UO_2. Moreover, Guerin recently measured the yield stress by using a compression method and obtained similar results at low stress ($5<\sigma<10$ kg/mm^2)(7).

In the second mechanism, the yield strength is controlled by the dislocation-fine scale prosity interaction or the dislocation-impurity atom interaction. Roberts et al. measured the effect of porosity on the strength of UO_2 and showed the increases of the yield strength with increasing porosity is caused by the increase of obstacles in the dislocation movement.

Finally, the yield strength can be controlled by the Peierls stress(3) (5) (11). Nadeau measured the dependence of the yield strength of UO_2 single crystal on the stoichiometry, and concluded the rate-controlling deformation mechanism for UO_2 may be the thermally activated surmounting of the Peierls barrier.

In the present work , the value of the activation energies greater than the one for uranium self-diffusion were obtained at the high strain rate, as shown in Table 2, and showed that the yield flow of UO_2 at high strain rate occurs in non-diffusion controlled mechanism.

Many workers(13)~(15) have investigated slow rate deformation in UO_2 and pointed out that the strain rate sensitivity ($\Delta ln\dot{\varepsilon}/\Delta ln\sigma$) is 4~5 when the deformation is controlled by the diffusion of uranium ion. However, the strain rate sensitivities in the present work were 14~33 in the temperature range and show disagreement with the values predicted from the diffusion mechanism.

In addition, grain-boundary sliding was not observed in the UO_2 specimens strained up to the yield stress level, but the slip lines were observed as shown in Photo. 1. These facts show that the grain-boundary sliding may not cause the yileding.

The yielding is generally sharp in appearence and its stress is little effected by strain rate(16), when the yielding is controlled by the dislocation-fine scale obstacle interaction. However, all the yielding observed in this work is smooth in the stress-strain curves and the yield stresses strongly depend on the strain rate. Therefore, the results rule out the possibility that the yield stress is not controlled by the dislocation-fine scale obstacle interaction. Roberts et al.(6) measured the elastic limit of UO_2 in the porosity range from 5 to 9 vol% and showed little porosity dependence on the elastic limit at 1,250° and 1,600°C. It is most likely that there is no effect of porosity on the yield stress in the present data, because the porosity of the specimens in this work is 6±1 vol% .

When crystals contain dislocations, their strength is governed by the transport properties and interactions of the dislocations(17). Especially, in ionic crystals, their strength is controlled primarily by the Peierls stress which arises from ions of the same sign having to pass close to one another during slip(11). If the motions of the dislocations are rate-controlled by overcoming the Peierls stress, it is not expected that the activation energy of the yielding is directly dependent on the strain [18].

Concurrent with the above mentioned fact, the activation energies of the yielding obtained in this work, are independent of the strain as shown in Fig. 7. Kurosawa[19] theoreticaly calculated the Peierls stress in ionic crystal, and showed that its value ranged from $2\times10^{-3}G$ to $6\times10^{-3}G$, where G is the shear modulus. The value[20] [21] of the shear modulus for UO_2 is approximately 6,900 (at 900°C)~6,000 kg/mm^2 (at 500°C), and thus the theoretical Peierls stress is 10~40 kg/mm^2 in the temperature range from 900° to 1,500°C. The yield stress obtained in this work agree with the theoretical Peierls stress in UO_2. Therefore, it is most likely that the yield stress of UO_2 at high strain rate is controlled primarily by the Peierls stress.

V. CONCLUSION

Polycrystalline UO_2 was strained in compression at constant high strain rates from 0.05 to 10.0 /min and at temperatures from 900° to 2,000°C. The following results are drawn:

(1) Polycrystalline UO_2 showed a characteristic yielding in the present study similar to the latest investigation of Guerin.

(2) The yield stress of UO_2 has strong temperature and strain rate sensitivities.

(3) The yield strength at high strain rate may be controlled primarily by the Peierls stress.

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Table 1 Uranium dioxide analysis

O/U ratio : 2.00±0.01
Density : 94±1% theoretical
Grain diameter : ~17μ
Impurity content :

Element	ppm on U basis
Al	<14
B	0.2
C	18
Ca	<10
Cd	< 0.3
Cl	<10
Cr	< 8
F	<10
Fe	70
Mg	6
N	21
Ni	10
V	< 3

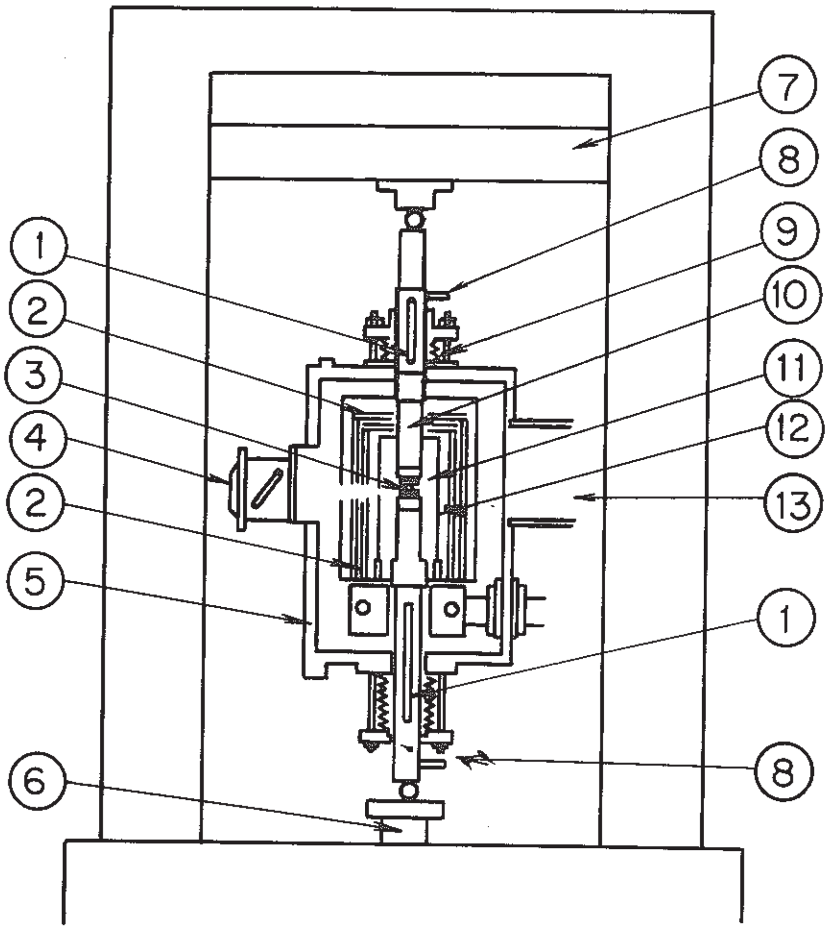
Table 2 Activation parameters of yielding for UO_2

Temp. (°C)	$(\Delta\sigma/\Delta T)_{\dot{\varepsilon}}$ (kg/mm²/°K)	$(\Delta \ln\dot{\varepsilon}/\Delta\sigma)_T$ (1/kg/mm²)	m ($\Delta \ln\dot{\varepsilon}/\Delta \ln\sigma$)	H (ev/atom)	H* (ev/atom)
1100	-0.053	0.965	23.7	8.3	5.0
1200	-0.034	1.370	23.3	8.7	4.0
1400	-0.013	2.421	33.3	7.6	3.0
1600	-0.016	1.546	28.7	7.5	2.5
1800	-0.023	1.342	13.7	11.4	2.5

$H = -kT^2 (\Delta\sigma/\Delta T)_{\dot{\varepsilon}} \cdot (\Delta \ln\dot{\varepsilon}/\Delta\sigma)_T$

m : Strain rate sensitivity

H* : Byron's data(2)



(1) Water cooled heat resistant steel, (2) Tantalum radiation shield, (3) Specimen, (4) Observation port, (5) Water jacket, (6) Load cell, (7) Cross head, (8) Coolant water, (9) Stainless steel bellows, (10) Heat-resistant steel, (11) Tungsten plates, (12) Tantalum element, (13) Vacuum line,

Fig. 1 Schematic diagram of high temperature compression testing apparatus

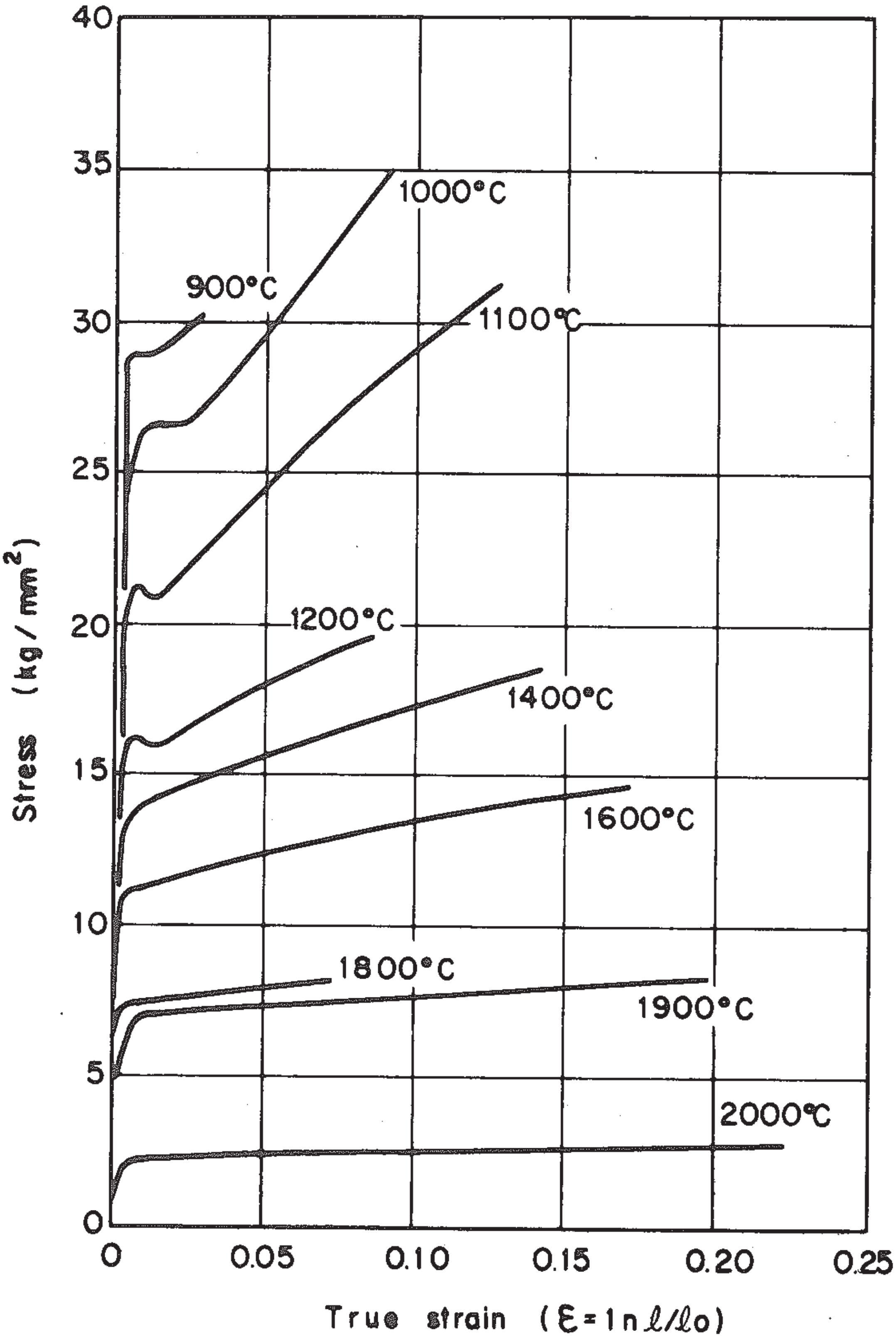


Fig. 2 Typical compressive stress-strain curves for UO_2
(Strain rate, $\dot{\varepsilon}$ = 0.1 / min)

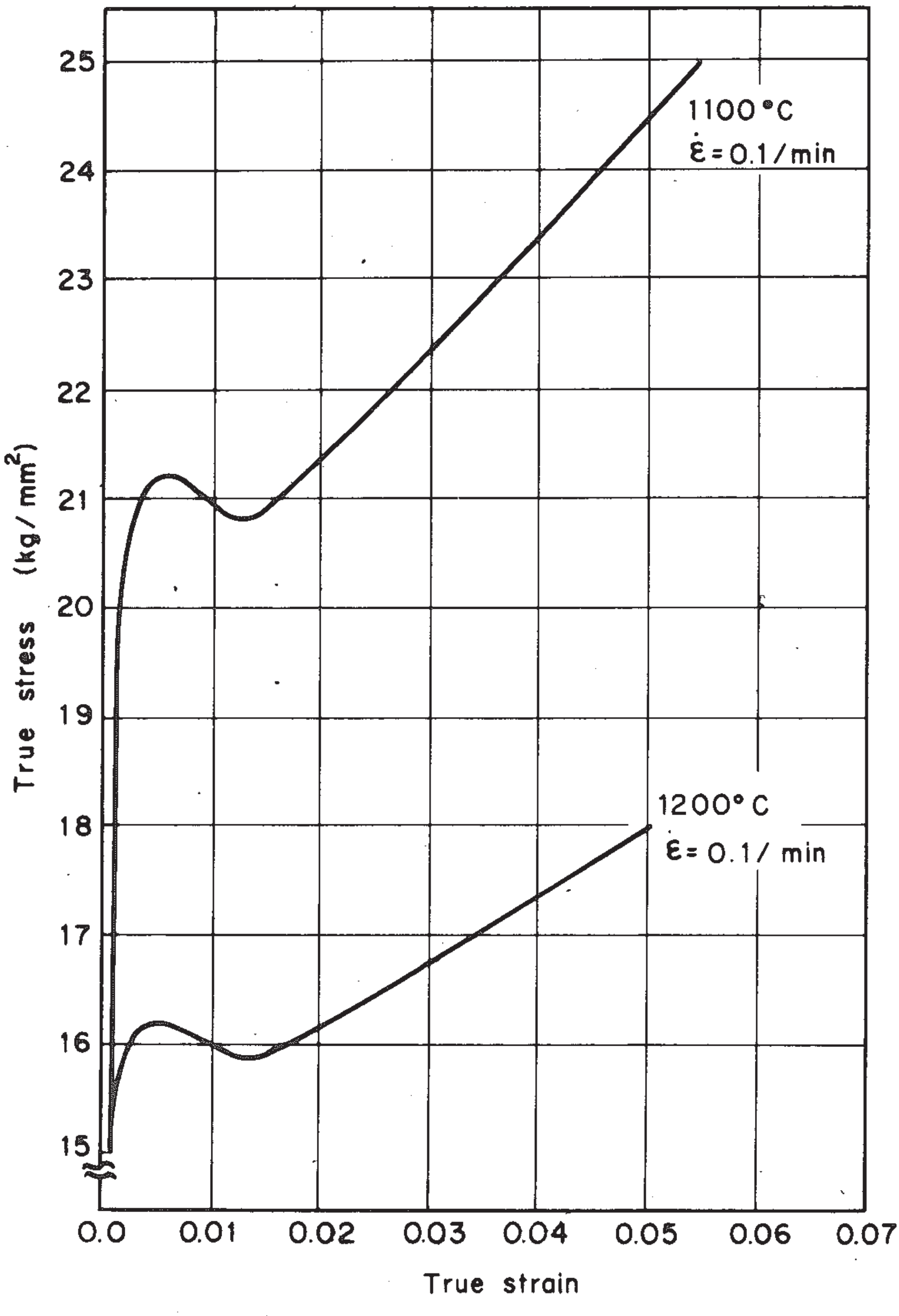


Fig. 3 Typical yieldings in UO_2.

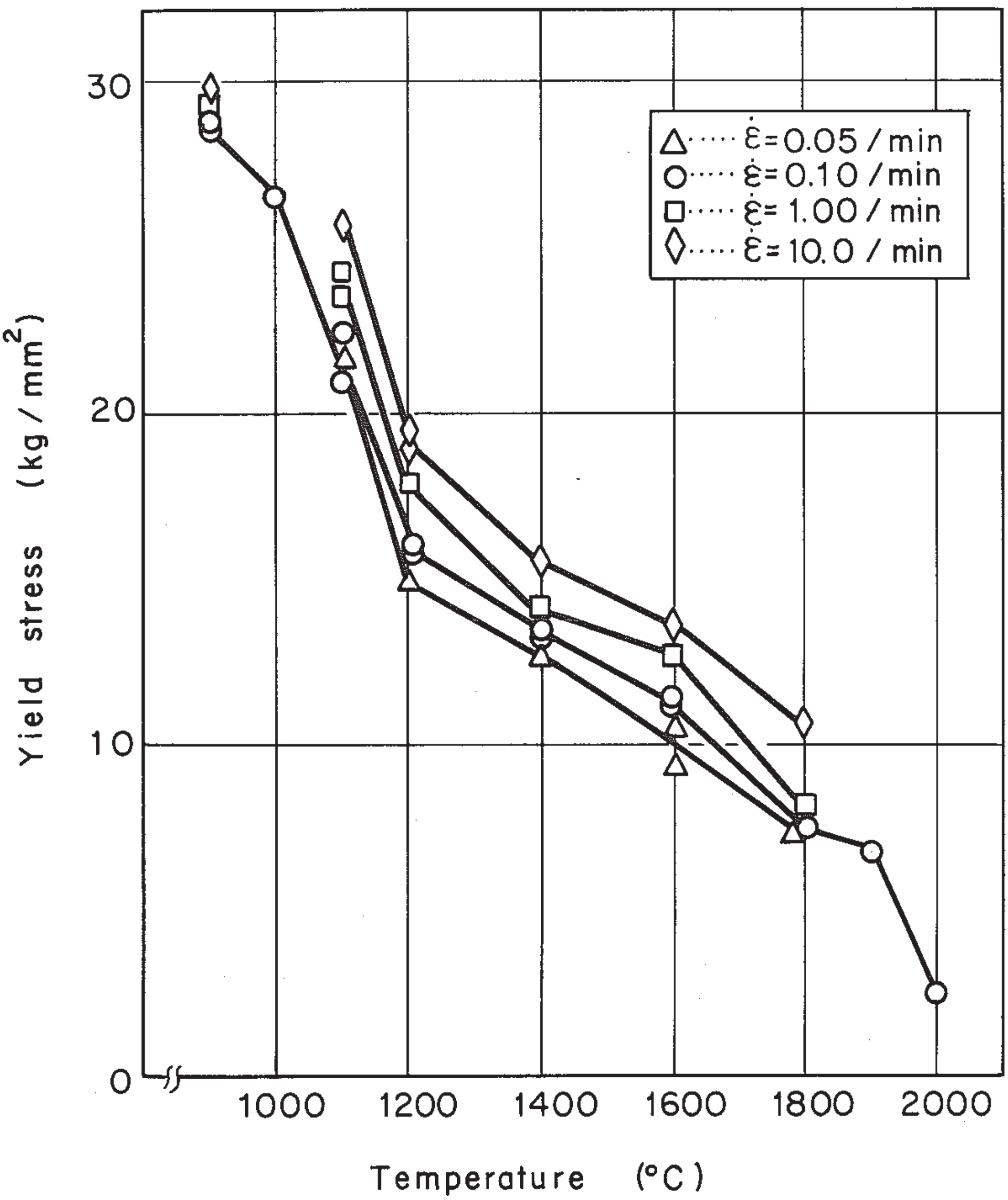


Fig. 4 Effect of temperature on the yield stress of UO_2

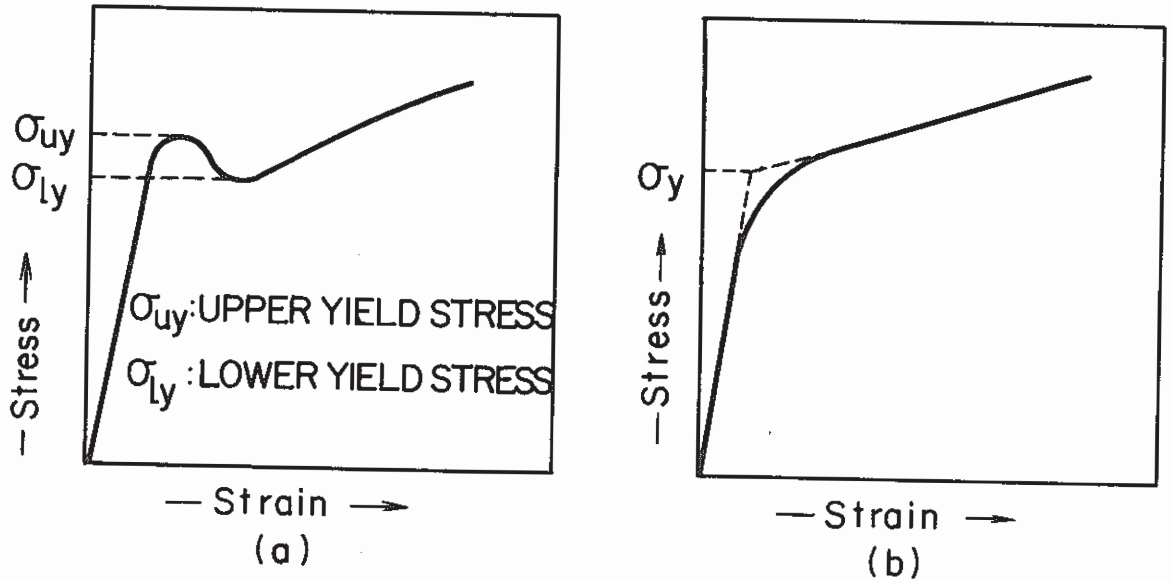


Fig. 5 Determination of yield stress

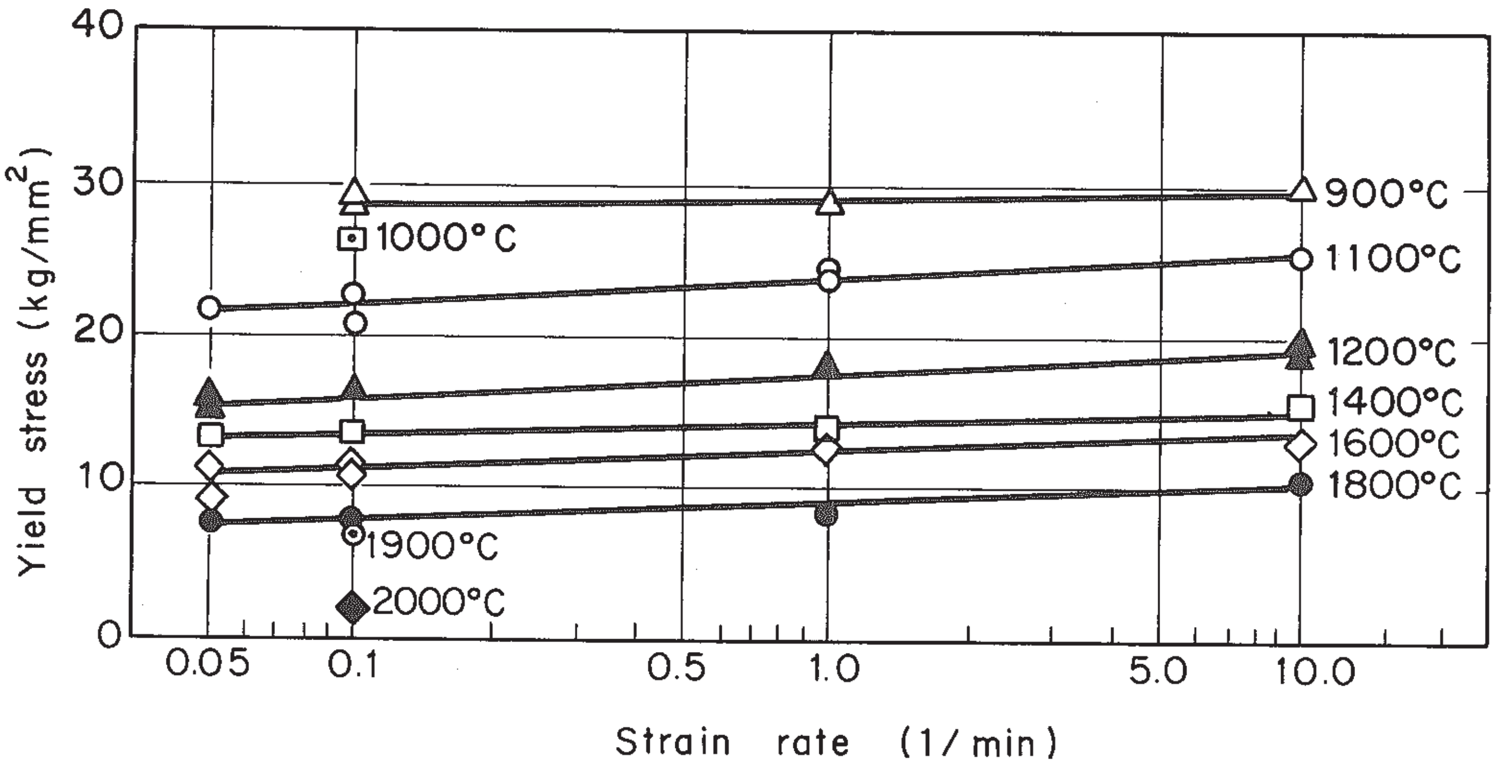


Fig. 6 Variation of yield stress in UO with the logarithm of the strain rate.

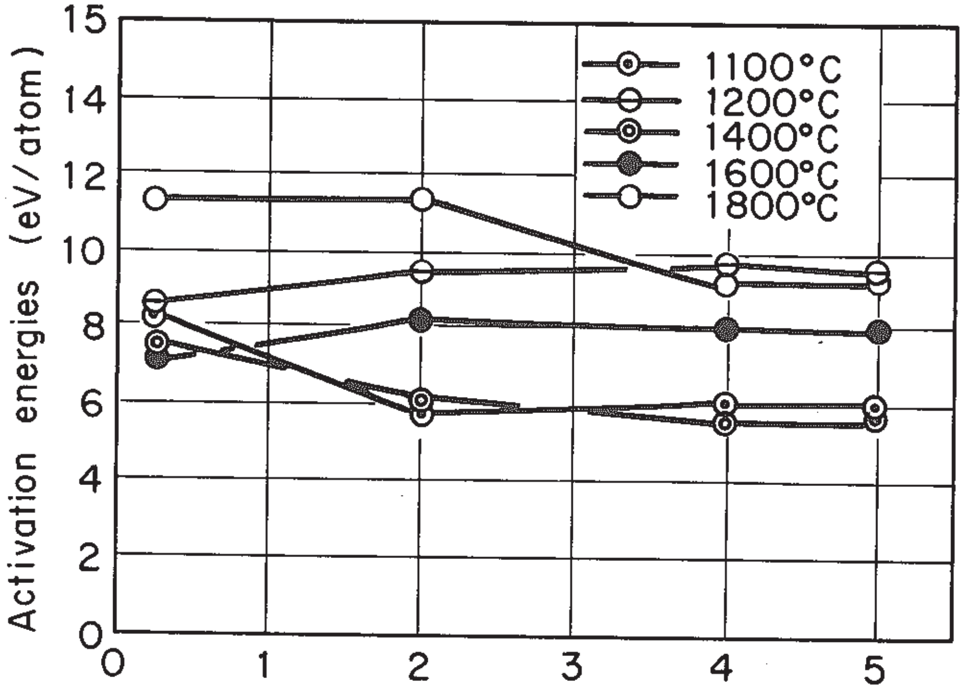


Fig.7 Effect of strain on the activation energies for plastic flow in UO_2.

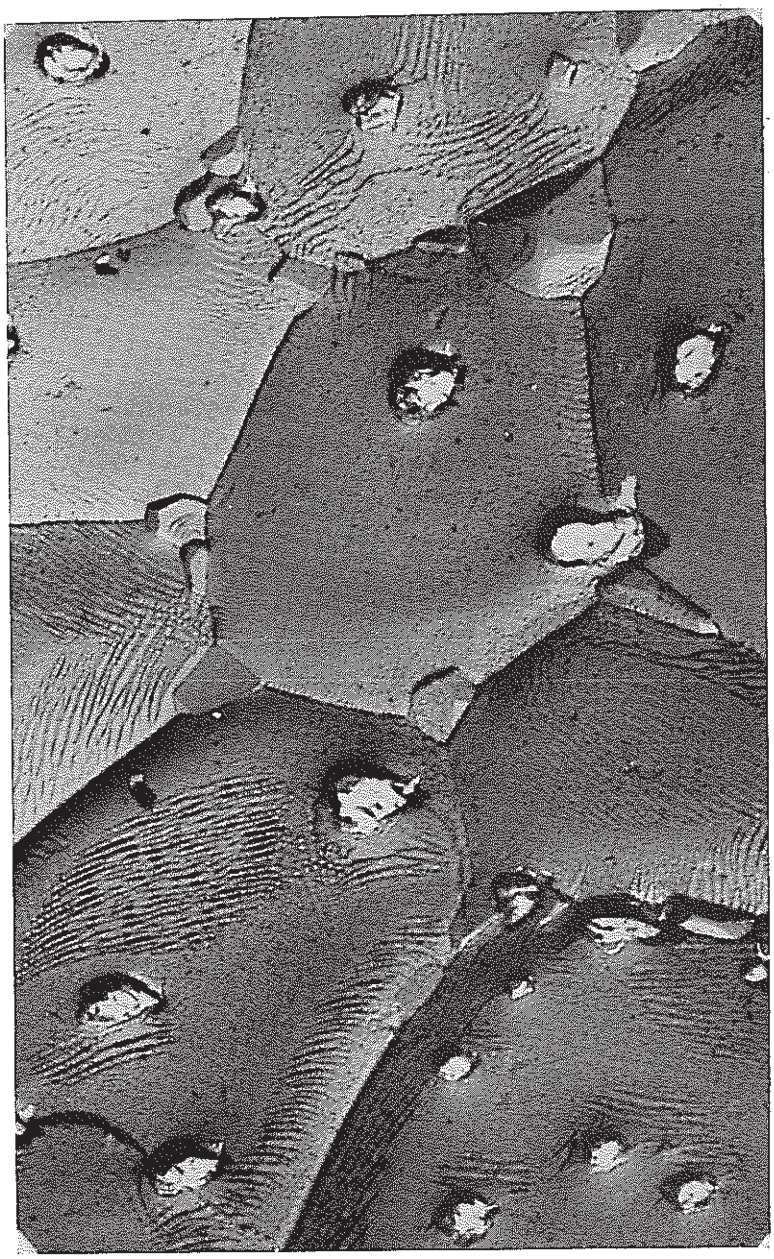


Photo.1 Electron micrograph showing the slip Lines in UO_2 deformed 2% in compression at 1600°C. The direction of compressive load in this test was parallel to the paper (x10,000).