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COMPRESSIVE CREEP OF ULTRAFINE GRAINED MgO POLYCRYSTALS

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Résumé. — Le fluage stationnaire de l'oxyde de Magnésium polycristallin à grains ultra-fins a été étudié entre 0,3 et 0,38 Tm. Les échantillons ont été fabriqués par frittage réactif durant la réaction de décomposition de $Mg(OH)_2$. Ce type de frittage produit un matériau dense (90 % de la densité théorique) possédant une taille de grains très fine (1 000 Å). La sensibilité de la vitesse de fluage stationnaire avec la contrainte s'exprime par un exposant de contrainte de l'ordre de l'unité et l'enthalpie d'activation est faible de l'ordre de 36 kcal/mole.

Abstract. — Steady state compressive creep of ultrafine-grained MgO has been studied in the temperature range 0.3 to 0.38 Tm. The specimens were fabricated by reactive hot pressing i.e. by hot-pressing of $Mg(OH)_2$ during the decomposition reaction, which produced dense specimens (90 % of the theoretical density) with 1 000 Å average grain size. The value of the stress exponent was found to be unity and the activation enthalpy was of the order of 36 kcal/mole.

1. **Introduction.** — It has been known for a long time that among the high temperature creep mechanisms (> 0.3 Tm) in ceramic polycrystals, those which are influenced by grain boundaries (i.e. Nabarro-Herring and Coble creep, or the more recent Ashby-Verrall mechanism) can contribute strongly to the deformation of these materials. On the other hand, another type of mechanism i.e. dislocation creep may also control the deformation in a significant way. As a matter of fact, it is often difficult to separate experimentally each of these mechanisms as they strongly interfere with one another in normal ceramic materials. Nevertheless, in ultrafine-grained ceramics, the possibility exists of observing a predominantly grain-boundary controlled creep behaviour. Thus, a large change in mechanical behaviour was observed in MgO having a grain size below 5 μm, and this has been associated with the onset of grain boundary sliding (Passmore, Duff and Vasilos 1965, Morgan 1970).

An ultrafine-grained ($\simeq$ 0.1 μm), almost fully dense (90 %) sintered MgO has been produced by reactive hot pressing. The purpose of the present paper is to describe this type of process, the characterization of the product and finally some preliminary mechanical data. The tests show that steady state creep occurs in the material at temperatures as low as 700 °C, with a low activation enthalpy.

2. **Experimental procedure.** — 2.1 SPECIMEN FABRICATION. — Discs of polycrystalline MgO, 20 mm in diameter and 6 mm thick, were produced by reactive hot pressing during the dehydroxylation of Magnesium hydroxide powder (Chaklader and McKenzie 1964). A pressure of 110 MPa was first applied at room temperature, and after mechanical equilibrium was attained the powder was then heated to 900 °C, while under pressure and maintained at this temperature for fifteen minutes for completion of the reaction. The specimen was then furnace cooled and no annealing treatment was performed. The almost fully dense specimen obtained had 96 % (wt %) purity. X-ray, thermogravimetric and infra-red studies indicated that there was $\sim$ 4 wt % $Mg(OH)_2$ in the system. The crystallite size of the disc was determined by the X-ray line broadening method, using the Scherrer formula (Scherrer 1918).

$$D = \frac{K\lambda}{\beta^{1/2} \cos \theta}$$

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where the half intensity width $\beta^{1/2}$ was further corrected for the instrumental width using the Warren equation (Warren 1941).

Line broadening calculations yielded an average crystallite size of about 1 000 Å.

2.2 SAMPLE PREPARATION AND MICROSTRUCTURAL EXAMINATION. — Creep samples were cut from the pressed disc with a boron carbide saw into $2 \times 2 \times 5$ mm^3 parallelopipeds. The density of samples was determined before and after creep using a hydrostatic weighing technique with alcohol. Before creep this density was 90 % of the *theoretical* density. In order to determine whether the sample density and the microstructure could be affected by heating alone, i.e. with no applied stress, another sample was laid unstressed near to the stressed sample during each creep test.

Microstructures of samples have been observed before and after creep tests by microscopic examination of freshly fractured surfaces, using either scanning electron microscopy, or carbon replicas, or thin foil transmission electron microscopy.

2.3 MECHANICAL TESTING. — Compression creep tests were conducted in air throughout the temperature range 700-860 °C, using constant stress equipment described elsewhere (Doukhan, Duclos and Escaig 1973). The stress dependence of the creep rate in the steady state region was determined using an incremental stress method which consisted of stress jumps performed on the same specimen while it was creeping at a steady rate.

The apparent activation enthalpy for creep was determined using conventional tests i.e. by plotting the steady state creep rate (in log scale) versus T^{-1} for different samples.

3. **Experimental results and discussion.** — Only preliminary results are reported below. Specimens were crept in the temperature range 700-860 °C under a constant applied stress of 140 MPa. Due to the small creep rate obtained, creep run durations were of the order of 48 hours.

3.1 CREEP LAW. — Figure 1 shows a typical creep curve obtained at $T = 816$ °C, and stress $\sigma = 140$ MPa. After a few hours of transient, a quasi steady state creep was reached ; this creep rate obeys the usual equation :

$$\dot{\varepsilon} = A\sigma^n \exp \frac{-\Delta H}{kT}$$

where A and n are constants, ΔH is the creep activation enthalpy, and K, T, and σ have their usual meaning. In this equation, the grain size dependence is not explicitly stated as samples of about the same grain

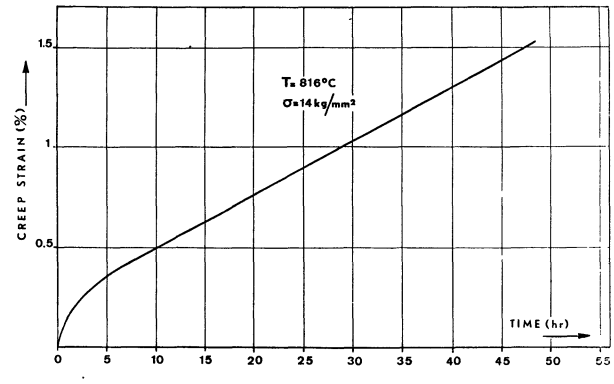

FIG. 1. — Example of a creep curve at a temperature $T = 816$ °C and under a stress $\sigma = 140$ MPa.

size were used in this study. The apparent creep activation enthalpy :

$$\Delta H_{app} = -kT^2 \left[\frac{\partial \operatorname{Log} \dot{\varepsilon}}{\partial T}\right]_{\sigma,\ \text{structure}} \simeq$$

$$\simeq k \frac{\operatorname{Log} \dot{\varepsilon}_2 - \operatorname{Log} \dot{\varepsilon}_1}{T_1^{-1} - T_2^{-1}}$$

is determined from creep rates observed at two different temperatures, under the same stress and same creep substructure. Figure 2 shows that a straight line results from plotting the steady state creep rate versus T^{-1} for different samples ; the slope gives :

$$\Delta H_{app} \approx 36 \text{ kcal/mole} .$$

The stress exponent :

$$n = \left[\frac{\partial \operatorname{Log} \dot{\varepsilon}}{\partial \operatorname{Log} \sigma}\right]_{T,\ \text{structure}} \simeq \frac{\operatorname{Log} \dot{\varepsilon}_2 - \operatorname{Log} \dot{\varepsilon}_1}{\operatorname{Log} \sigma_2 - \operatorname{Log} \sigma_1}$$

is measured from differential tests. Stress jumps ($\Delta\sigma \simeq 0.13\ \sigma$) give an approximate value $n \simeq 1$ in the whole temperature range.

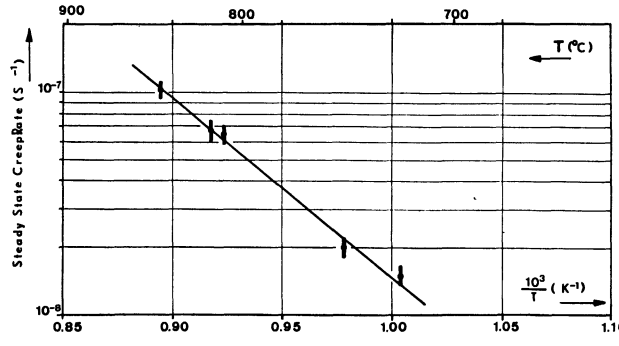

FIG. 2. — Creep rate (Logarithmic scale) versus T^{-1}, for the constant stress $\sigma = 140$ MPa, and the constant grain size $d = 1\ 000$ Å.

3.2 CREEP MICROSTRUCTURE. — Figure 3 shows a representative T.E.M. photomicrograph of samples before testing. Individual cristallites are revealed by the dark field technique. The polycrystalline MgO

FIG. 3. — 100 kV electron micrograph of as pressed sample; dark field condition.

obtained by reactive hot pressing at 900 °C was ultra-fine grained and rather uniform in texture with grain size distribution averaging 1 000 Å. In several foils, it was observed that grains did not contain dislocations. Some evidence of the pore phase (~ 10 %) could be observed. The pore size was less than the grain size. The pores were generally dispersed throughout the matrix in an inter-granular way, but a channeling distribution could be sometimes observed.

The S.E.M. fractograph (Fig. 4) illustrates the microstructure as revealed by fracturing uncrept, and unheated samples at room temperature. Essentially intergranular fracture was observed.

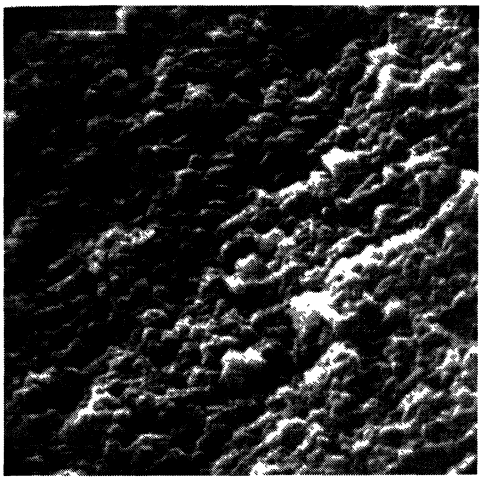

FIG. 4. — S.E.M. fractograph of as pressed sample.

No detectable grain growth occurred at test temperatures for the crept or for the uncrept samples as shown in figure 5, which was replicated from freshly fractured surface of specimens crept at 720 °C and 860 °C to about 1.5 % strain.

Finally no decrease or increase in bulk density was detected during heating and creep tests.

3.3 DISCUSSION AND CONCLUSION. — It is possible by reactive hot pressing to produce almost fully dense polycrystalline MgO with an ultra-fine grain size

FIG. 5*a*. — Carbon replica electron fractograph of 1.5 % creep strained sample at 720 °C under a stress σ = 140 MPa.

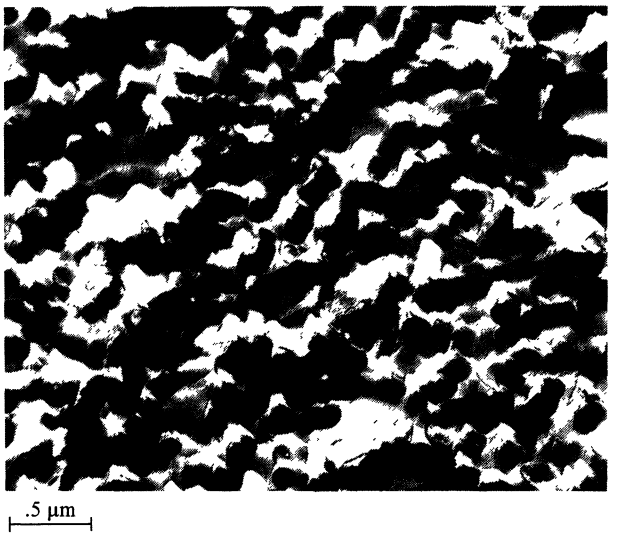

FIG. 5*b*. — Carbon replica electron fractograph, sample crept at 1.5 % at 860 °C, under a stress σ = 140 MPa.

(1 000 Å). This material is a ductile ceramic when crept at high temperatures. At 850 °C and under a stress of 140 MPa, its creep rate $\dot{\varepsilon}$ is of the order of 10^{-7} s^{-1}.

The above data obtained from creep experiments on a relatively small number of specimens are not considered adequate for a quantitative statement of the values of n and ΔH. The trends are however now clear. The stress exponent is small, of the order of unity, and the apparent creep activation enthalpy is low, about 36 kcal/mole.

Microstructural observations showed that the retention of the initial structural properties (grain size, no dislocations in grain, density) after a small creep strain $\leqslant$ 2 %.

These preliminary investigations indicate that deformation mechanism may be boundary-diffusion controlled in this ultra-fine-grained polycristalline MgO.

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