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Anelastic Grain-Boundary Sliding in Ceramics. I. Directly Bonded Polycrystals

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Abstract. Internal friction of high-purity single-crystal sapphire and three polycrystalline aluminas with different grain sizes has been measured up to very high temperature at the frequency of 10 Hz. Only an exponential background curve was found in the single-crystal while polycrystalline specimens showed a broad anelastic peak, superposed on the background component, whose nature was presumably related to the presence of internal grain boundaries. The overall internal friction curve of the polycrystalline specimens were markedly shifted to lower temperatures as compared to sapphire. The intensity of the grain-boundary peak was decreasing with increasing the grain size. A series of alloys, prepared by doping with increasing amounts of Cr_2O_3 , were also tested in comparison with ruby single-crystals. The Cr addition led to both an increase in intensity of the grain-boundary peak and a shift of its maximum towards lower temperatures. This characteristic anelastic behavior was clearly dictated by a lowered coherency of grain boundaries upon Cr alloying.

1. INTRODUCTION

Internal friction technique is one of the few experimental tools capable to provide direct information about the microscopic diffusive processes occurring either at grain boundaries or within the grains of polycrystalline solids upon increasing temperature. So far, internal friction measurements have been widely and successfully applied to analyze the behavior of both metals and related alloys [1]. Several internal friction studies have also been reported for ceramic polycrystals, mainly in the attempt to phenomenologically relate the damping characteristics to the various processes of strength degradation occurring in polycrystalline solids at high temperature [2,3]. However, only a very few evidences have been so far provided about grain-boundary-related internal friction response of polycrystalline ceramics mainly because of the rather high range of temperature needed for performing a meaningful internal friction analysis. One critical issue on the topic also deals with the difficulty in modifying the bond between grains without changing their morphology and bulk deformation behavior.

In this paper we deal with high-purity alumina phase whose boundaries were found to be free from impurities (i.e., directly bonded) up to the resolution allowed by the transmission electron microscope, namely 0.18 nm [4]. It was early recognized that high-purity alumina ceramic with "clean" grain boundary would be an interesting candidate for basic internal friction investigations [5]. This is because it would be relatively easy to obtain significant grain growth without addition of extraneous phases or additives. Furthermore, alumina ceramics undergo a wide range of known solid-solutions which may modify the degree of bonding and coherency of the grain-boundaries. Both this characteristics are examined in the present study.

2. EXPERIMENTAL PROCEDURE

Historically, two methods of investigation have been described in the metal field; the first, due to Kê [6], is the measurement of the anelastic behavior of specimens differing only in the grain boundary area. In other words, upon the occurrence of significant grain growth process, the strength of the anelastic grain-boundary peak was found to clearly decrease until disappearing when the grain size was comparable

with the specimen size or in single crystals. Another approach, widely followed in metals [1], deals with the modification of the grain-boundary bonding through the occurrence of solid solutions.

Four kinds of high-purity aluminium oxide specimens were first tested. They were: three polycrystalline aluminas with different average grain size and one single-crystal sapphire. The polycrystalline specimens were pressureless sintered by the authors using high-purity alumina as a starting powder (Sumitomo AKP-53). Sintering up to full densification was conducted in air with holding the specimens at 1873K for 3 hours. Iterative annealing cycles were conducted at 1773K for 2–10 hours in order to produce appreciable grain growth in the polycrystalline body. The average grain sizes of the polycrystalline specimens were found to be 3.3 (in the as-sintered status), 4.8 (for 2 h annealing) and 19.3 μm (for 10 h annealing). One large, high-purity (>99.99%) single-crystal sapphire rod was purchased from Nakazumi Crystal Co. and cut in suitable dimensions with the *c*-axis of the crystal nearly parallel to the specimen axis (i.e., the axis of torsion). Annealing for two hours at 1973K was then performed in order to release eventual residual stresses induced by machining.

Polycrystalline alumina specimens containing 0.1, 0.5, 1.0 and 5.0 wt% Cr, respectively, were also prepared by hot pressing at temperatures in the range 1723–1823K for 2 h, under 30 MPa. The hot-pressing temperature was empirically optimized in order to achieve nearly full density and a close grain size and morphology (i.e., equiaxed grains 4–6 μm in size) in the four specimens. For comparison, also two ruby crystals were tested. Their Cr content was 0.3 and 3.0 wt%, respectively. The annealing procedure was the same as that described for the sapphire crystal.

Internal friction data were automatically collected at intervals of 5K, according to the free-decay method [1]. Experiments were conducted at the fixed frequency of 10 Hz. The maximum applied torsional stress was ≈ 10 MPa. In order to achieve very high temperatures, the torsional pendulum apparatus was equipped with a carbon heater, rolled around the specimen, and an infrared thermo-analyzer [7]. Preliminary blank-test calibrations by strain gauges were performed to minimize the residual strains in the fixed bottom grip and the extension shaft of the pendulum. Internal friction could be ultimately measured up to the melting point of sapphire, found to be in agreement with the value $\approx 2323\text{K}$ reported in previous literature [8].

3. RESULTS AND DISCUSSION

Figure 1 shows the internal friction curves of high-purity alumina polycrystals with various grain sizes, *d*, as a function of the absolute temperature, *T*. For comparison, also the internal friction curve measured in single-crystal sapphire is reported. Very small, if any, relaxation peak was found in the sapphire rod, its damping curve being simply represented by an exponential-like background. The background curve of sapphire was rising very slowly up to about 10K below the melting point, above which a steep rise occurred. The curves of the polycrystals resulted markedly shifted towards lower temperatures, the lower the temperature, the smaller the grain size. It could be argued that the anelastic response of the specimen was directly related to the amount of grain-boundary surface area present in the specimen. Focussing only on the phenomena related to the presence of grain boundaries, the exponential (plastic) background curve found for the sapphire rod can be subtracted from those of the polycrystals. It was thought that, by doing so, the remaining component of internal friction of the polycrystals would represent the anelastic contribution of grain boundaries. A broad peak is revealed by such a procedure as shown, for example, for the polycrystalline alumina with smaller grain size in Fig.2. It is interestingly found that the anelastic peak tends to disappear at larger values of grain size. This evidence, consistent with early studies by Ke [5] on pure aluminum, appears to provide experimental foundation for relating the origin of the peak to slip at grain boundaries of alumina ceramics.

Figure 3 shows the internal friction curves of two ruby single-crystals and a series of Cr-doped alumina polycrystals as a function of the absolute temperature. For an easier comparison, the data for the sapphire specimen from Fig.1 are also replotted. Again no peak was clearly detected in the ruby single crystals, the background curves rising quite slowly up to very high temperature. The temperature from which the steep rise took place was function of the Cr content, the higher the content the higher the temperature. In the polycrystalline specimens, the increasing additions of Cr have the double effect of shifting the internal friction curve towards lower temperatures and increasing its absolute value as compared to the pure alumina polycrystal with the same grain size. The former characteristic cannot be dictated by the bulk behavior of the grains, since it reflects exactly the opposite trend as found for the single crystal specimens. Also, any diffusional (i.e., intragranular) origin of the peak is to be *a priori* excluded since, otherwise, we should be able to monitor it also in the single crystals. Hence, the evolution of the internal friction curve upon Cr addition should be related to a somehow modified structure of the grain boundary. The relaxation peaks, as a function of Cr content, are shown in Fig.4 after subtracting the respective exponential backgrounds. The effect of solid-solution by Cr, as discussed above, is best seen in this figure. The apparent activation energy for the present anelastic relaxation peak de-

creases as the material becomes more Cr-rich. The apparent activation energy values, as calculated from the peak width method [1], were found to be 243, 234, 176 and 121 kJ/mol for the 0.1, 0.5, 1.0 and 5.0 wt% Cr-doped polycrystals, respectively. Although the present activation energy values may be underestimated because of broadening of the peak, the value found for low Cr-doped material is close to that reported for grain-boundary diffusion in pure alumina polycrystals [9]. Read and Shockley [10] have proposed that, in metallic materials, the unit mechanism in the process of stress relaxation across a directly bonded grain boundary is the transfer of atoms from one edge dislocation to another. In such a process the generation of a vacancy at an edge dislocation results to be the rate-limiting process. Unfortunately, the present data do not allow us to judge whether or not this theory is directly applicable to the present ceramic materials. Furthermore, it is not straightforward how an isovalent solute like as Cr in alumina can ease vacancy generation. From the present data, it seems to follow that the increase of nominal amount of Cr atoms in the material involves easier sliding of the grain boundaries. In fact, both the higher intensity and the increased broadness of the peak as well as its lowered peak-top temperature (cf. Fig. 4) suggest that grain-boundary sliding occurs in a statistically higher number of boundaries when Cr content increases. In absence of a glassy phase of finite thickness at the grain boundary [3], the present results would suggest that the presence of Cr may lead to a modified structure, namely to a lower coherency, of grain boundaries in alumina ceramics.

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Figure 1: Internal friction curves of single-crystal sapphire and three polycrystalline aluminas with different grain sizes.

Figure 2: Broad grain-boundary relaxation peak for pure polycrystalline alumina with fine grain size, as revealed by subtracting the exponential-like (plastic) background of the sapphire specimen.

Figure 3: Internal friction curves of ruby single crystals and polycrystalline alumina-Cr solid solutions as a function of temperature. The curves were obtained at the frequency of ≈ 10 Hz.

Figure 4: Evolution of grain-boundary relaxation peak in polycrystalline alumina upon increasing the amount of Cr in solid solution.