Creep of hot-pressed silicon nitride ceramics

Although hot-pressed silicon nitride ceramics exhibit high strengths at room temperature, the deformation resistance of this type of material decreases rapidly at elevated temperatures. This strength degradation is influenced by the presence of a glassy silicate phase at β-Si_3N_4 grain boundaries which is formed by reaction between the hot-pressing additive (usually MgO) and an oxidized (SiO_2) layer at the surface of the silicon nitride particles. Since both slip and diffusional processes are difficult with this group of materials, the high-temperature creep properties have generally been interpreted in terms of deformation processes dependent on the presence of the intergranular silicate phase. These include grain boundary sliding controlled by the viscosity of the glassy phase [1], dissolution and reprecipitation of material [2] and transfer of the viscous phase from boundaries under compression to those under tension [3]. All of these mechanisms are related to the viscosity of the grain boundary silicate, accounting for the dependence of the creep resistance on factors such as the type and level of impurities, particularly CaO [1, 2, 4]. However, with hot-pressed silicon nitride, a compressive stress of 689 $MN\,m^{-2}$ was required to obtain a creep rate of $\sim 1.5 \times 10^{-7}\ sec^{-1}$ at 1643 K which was more than ten times the stress needed to produce this creep rate under tensile creep conditions [1]. With processes dependent on the viscosity of the grain boundary phase, comparable creep rates would be expected in tension and compression. It has therefore been suggested that the rate-determining process during creep is not sliding *per se* but the accommodation of sliding by grain boundary microcrack development [5]. In a compression test, the maximum tensile stresses generated are only about a tenth of the applied compressive stress. Intergranular crack formation depends on the tensile stress level across grain boundaries so that the greater strength in compression would therefore be expected.

In a recent study of the compression creep characteristics of a wide range of silicon nitride ceramics, tests were carried out on two hot-pressed products having different levels of MgO additive [6]. The sample containing ~2% MgO was close to theoretical density ($3.2 \times 10^3\ kg\,m^{-3}$) and was composed almost entirely of β-Si_3N_4 with a crystal size of ~1 μm. The creep strength of this material was considerably greater than that for test-pieces having comparable density and crystal size, but which contained ~5% MgO (Figs. 1 and 2). In the present programme, a further series of creep tests were undertaken in compression for samples from an experimental batch of hot-pressed material having ~2% Y_2O_3 rather than MgO as the pressing additive. Since the densities and crystal sizes of these materials were similar, an assessment can be made of the effects of variations in the composition and amount of grain boundary silicate on the creep behaviour of hot-pressed silicon nitride.

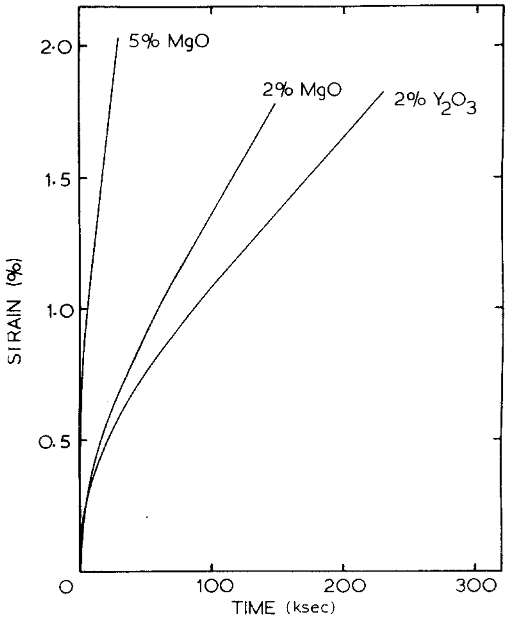

Figure 1 Creep curves for hot-pressed silicon nitride samples containing either 2% MgO, 5% MgO, or 2% Y_2O_3 as pressing additives. The tests were carried out at 238 $MN\,m^{-2}$ and 1623 K.

The constant-stress tests were carried out at 1623 K using the high-precision equipment used in the earlier study for the MgO-containing samples [6]. The creep strains and the secondary creep rates recorded for the Y_2O_3-bearing material were slightly lower than those obtained for the samples having ~2% MgO tested under comparable conditions (Fig. 1). However, the stress dependences of the creep rates were similar (Fig. 2). Furthermore, temperature-cycling experiments during secondary creep of the Y_2O_3-material gave values of the activation energy for creep (Q_c) which were

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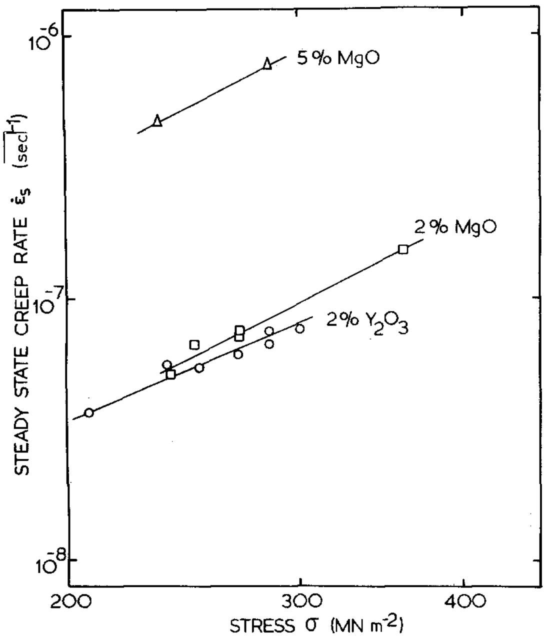

Figure 2 The variation of the steady state creep rate with applied stress for tests carried out at 1623 K for hot-pressed silicon nitride samples containing 2% MgO, 5% MgO or 2% Y_2O_3 as pressing additives.

within the range 650 (± 25) kJ mol^{-1} recorded for the samples produced using MgO [6]. The variation of the secondary creep rate, $\dot{\epsilon}_s$, with stress, σ, and temperature, T, for all of the hot-pressed materials studied can therefore be expressed as

$$\dot{\epsilon}_s = A\sigma^n \exp(-650\,000/RT)$$

with a stress exponent, n, of 2.1 to 2.3 [6].

Whilst the creep resistance of hot-pressed silicon nitride is clearly affected by the composition and amount of grain boundary silicate phase, the fact that the stress exponent and the activation energy for creep are similar for the three materials considered indicates that the deformation processes taking place are essentially the same. A stress exponent of ~2 is compatible with creep controlled either by the viscosity of the silicate phase [1–3] or by microcrack development [5]. The importance of viscous flow has been inferred from the observation that the activation energy for flow of certain silicate glasses [7] can be of the order of the activation energy for creep of HPSN (~650 kJ mol^{-1}). However, the activation energy for viscous flow of silicates varies with composition of the melt. The Q_c values would therefore be expected to depend on the amount and type of pressing additive if processes such as grain boundary sliding, viscous phase transfer, etc. determine the creep rate. The addition of ceria to HPSN, which increases the refractoriness of the grain boundary phase, has been reported to decrease the creep rate and increase the activation energy for creep [8]. Yet, the present results demonstrate that Q_c is independent not only of the amount of silicate phase but also of the composition. With creep controlled by microcrack formation [5], an activation energy which is essentially independent of the type and amount of pressing additive would be expected because, with the levels considered, the silicate phase is not continuous along the grain boundaries [9]. In addition, Q_c values of ~650 kJ mol^{-1} are close to those expected for crack growth of silicon nitride ceramics [6]. The results of the present investigation therefore suggest that creep of hot-pressed silicon nitride occurs as a result of grain boundary sliding controlled by microcrack formation, with factors such as the impurity levels modifying the creep resistance by affecting the ease of sliding and microcrack development.

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P. J. DIXON-STUBBS
B. WILSHIRE
Department of Metallurgy and
Materials Technology,
University College, Singleton Park,
Swansea, UK