THERMAL EXPANSION AND LINEAR-DIMENSION HYSTERESIS FOR POLYCRYSTALLINE SILICON CARBIDE

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Length measurements have been made on polycrystalline silicon carbide on cooling from 290 to 23 K, and the linear-expansion coefficient has been determined. The lack of reproducibility in the length after cooling has been examined. The irreproducibility after the first cooling is 10^{-5}, but after four cycles of cooling at low rates, the linear dimensions stabilize. The temperature dependence has been derived for the thermal relative length change in equilibrium silicon carbide specimens.

An optical system working over a wide temperature range needs materials with low thermal expansion coefficients, high thermal conductivity, good strength, and good elastic moduli. Silicon carbide has almost all those features, but no measurements have been made on certain physical properties for the polycrystalline material and the changes in them on thermal cycling. Major such characteristics are the thermal expansion and the effects of low-temperature cycling on the dimensions.

We examined the thermal expansion coefficient and the temperature irreproducibility in the linear dimensions on thermal cycling for polycrystalline silicon carbide at 20-300 K. For the single crystal, there are data [1] on the thermal expansion above 100 K, so the two forms could be compared. The irreproducibility on cooling can provide information on the thermal expansion in conjunction with the temperature dependence of the expansion coefficient and indicate the stability of items made from it.

The cylindrical rods 25 mm in diameter and 100 mm long were cut from a single batch of material and were examined in the state as supplied, not being given any special heat treatment.

The thermal expansion was measured on three specimens at 23-290 K by means of a capacitance dilatometer [2]; the error in determining the thermal expansion coefficient did not exceed $\pm 1 \cdot 10^{-7}$ K^{-1}.

Before the thermal expansion was measured, each specimen was checked for irreproducibility on thermal cycling. The initial length L_0 was recorded with the dilatometer at 280 K, and then the specimen was cooled along with the cell at about 10^{-3} Pa to 20 K at a rate of about 1 K/min. After the first cycle (cooling and heating to the initial temperature), the relative change in length $\Delta L/L_0$, i.e., the thermal irreproducibility, was usually about 10^{-5}, but even after the second cooling cycle, it was much reduced. Figure 1 shows the response to the number of cycles N for one of the specimens. The first cooling causes the specimen to extend, which is followed by a certain tendency to contract. After the fourth cycle, the length remains constant within the measurement error of $\pm 5 \cdot 10^{-7}$. The error in determining the irreproducibility was $\pm 2 \cdot 10^{-7}$.

These measurements show that one should determine the linear expansion coefficient α for polycrystalline silicon carbide after four cooling cycles. One measures $\alpha = \Delta L_T \cdot L^{-1} \cdot \Delta T^{-1}$ in the stationary state with discrete changes in temperature ΔT of from 3 to 10 K in accordance with the temperature range. Figure 2 shows the averaged $\alpha(T)$ curve from three specimens. The spread in the data for the different specimens did not exceed the error of measurement. The figure also shows published data [1] for single-crystal silicon carbide (1), which within the overall error agree with our values for the polycrystalline material. The temperature dependence is integrated to obtain the relative change in

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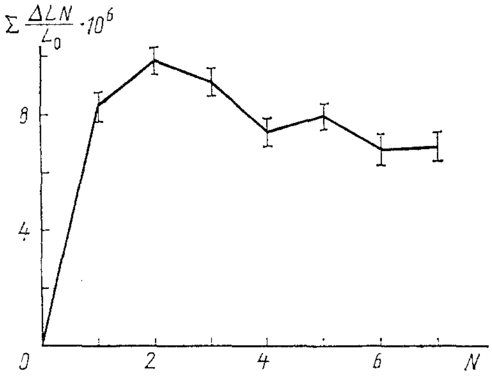

Fig. 1

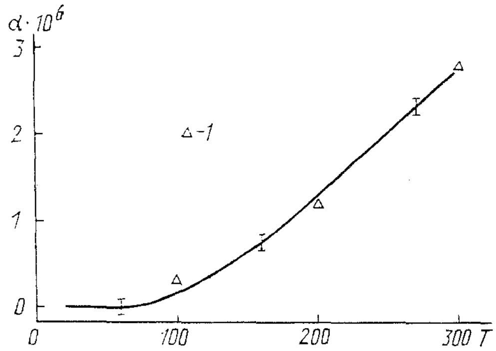

Fig. 2

Fig. 1. Effects of number of cycles N in the range 300-20 K on the overall temperature irreproducibility in the linear dimensions for polycrystalline silicon carbide.

Fig. 2. Temperature dependence of the linear expansion coefficient for polycrystalline silicon carbide (solid line): 1) [1] data for a single-crystal specimen, T in K, α in K^{-1}.

length $\Delta L_T/L_{300}$. Linear extrapolation was used with α(T) from 290 to 300 and from 23 to 20 K. Table 1 gives the calculated $\Delta L_T/L_{300}$, which can be explained from the history and microstructure. Polycrystalline silicon carbide is made by reactive sintering [3], which gives grains of the carbide in the main of the α phase with the presence of all polytypes. The grains are joined by secondary silicon carbide formed by combination of particles with silicon from the liquid and/or gaseous phase. The similarity in α for our material and the single crystal indicates that the thermal expansion is largely determined by the carbide grains. The somewhat reduced α at $T < 120$ K relative to the published values may be due to a small amount of free silicon, which has a negative expansion coefficient in that range.

Below 300 K, the anisotropy in the thermal expansion of α-SiC is slight [1], so the orientations of the SiC grains do not affect the expansion mechanism. Consequently, there are no causes of stresses or cracking on equilibrium cooling, and the length of the specimen should not alter on thermal cycling. That conclusion agrees with the asymptotic behavior of the irreproducibility in Fig. 1 (stresses naturally arise at high cooling rates).

The first cycle always causes the specimen to elongate, which is ascribed to the effects from the procedure for cooling from the sintering temperature, which attains 2000 K. Silicon carbide has considerable thermal-expansion anisotropy at high temperatures [1], so strains arise in the material because of the differences in shortening between the variously oriented grains, which result in residual strain and even in microcracks, which evidently occur mainly at low temperatures, when the specimen loses plasticity. As those processes are associated with thermal deformation, they should occur at temperatures where the thermal expansion is substantial. Table 1 shows that the expansion coefficient is very low below 100 K and there is virtually no thermal change in length. Therefore, the sizes of polycrystalline specimens can be stabilized by thermal cycling not down to 20 K but to 77 K, i.e., liquid nitrogen temperature.

However, Fig. 1 shows that a single cooling cycle is not sufficient for this. The residual stresses that are not released by cracking in the first cycle may make themselves felt in the second cycle and cause new cracks and slight stretching. At the same time, the microstructure should become ordered somewhat as the stresses are relieved and the microcracks are formed. The tendency to a more equilibrium microstructure reduces the size, as is observed in the third and fourth cycles. Naturally, microstructure ordering does not occur at constant temperature, and it is necessary to apply certain forces, which arise during thermal stressing from inhomogeneous temperature change.

The main conclusions are that the thermal expansion in technical polycrystalline or single-crystal silicon carbide is virtually the same for both at 100-300 K. Most of the relative length change of $2.80 \cdot 10^{-4}$ occurs at 100-300 K. On first cooling to 77 K and subsequent heating, the polycrystalline material shows irreproducibility of about 10^{-5} in

TABLE 1. Temperature Dependence of the Relative Length Change $\Delta L_T/L_{300}$ for a Polycrystalline Silicon Carbide Specimen

T, K	$\frac{\Delta L_T}{L_{300}}$, 10^{-4}	T, K	$\frac{\Delta L_T}{L_{300}}$, 10^{-4}
		160	−2,501
20	−2,823	170	−2,406
30	−2,822	180	−2,299
40	−2,821	190	−2,178
50	−2,820	200	−2,045
60	−2,819	210	−1,898
70	−2,818	220	−1,739
80	−2,812	230	−1,567
90	−2,810	240	−1,382
100	−2,798	250	−1,184
110	−2,778	260	−0,973
120	−2,747	270	−0,749
130	−2,704	280	−0,513
140	−2,649	290	−0,263
150	−2,582	300	0

size. The equilibrium state is provided by four cooling cycles to about 100 K with a rate of 1 K/min, after which the dimensions are stabilized to $\pm 5 \cdot 10^{-7}$.

These results lead one to recommend that polycrystalline silicon carbide should be used as a constructional material with low thermal expansion in the range from 20 to 300 K after those technological procedures.

NOTATION

L_0 and L_{300} initial length and length at 300 K; ΔL irreproducibility in length caused by cooling; N number of cooling cycles; ΔL_N irreproducibility in length after cooling cycle N; $\Sigma \Delta L_N/L_0$ relative overall thermal irreproducibility in length after N cooling cycles; ΔL_T temperature-dependent length change; α temperature coefficient of linear expansion.

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