Investigation of current-carrying capacity of bulky single-phase $PbMo_6S_8$ samples

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Results are presented of investigations of the critical currents and of the scaling laws for the pinning force in bulky single-phase $PbMo_6S_8$ samples with various grain sizes. It is shown that the critical-current density increases with decreasing grain size and reaches for samples with ≈ 0.3 μm grain size the relatively high value 3×10^8 A/m^2 in a field 10 T at 4.2 K.

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The record high values of the upper critical field B_{c2} in ternary molybdenum sulfides (TMS) of the type $PbMo_6S_8$ makes the question of the current-carrying capacity of superconducting materials based on TMS quite timely. The results of Refs. 1–4 offer evidence that in bulky sintered TMS samples the critical-current density J_c can reach values $(5–8)\times 10^8$ A/m^2 in weak fields and $(1–1.5)\times 10^8$ A/m^2 in fields ≈ 10 T. It is also shown in Refs. 4 and 5 that in such samples the functional dependence of the pinning force P_v on the relative induction $b = B/B_{c2}$ is described by general laws that are common to many known type-II superconductors with large current-carrying capacity. All these results were obtained with bulky sintered TMS samples prepared by direct synthesis from the basic components followed by homogenizing annealing of the sintered material compressed beforehand at high (≈ 30 kbar) pressure in cylindrical matrices. As a rule, substantially larger values of J_c were observed either when the content of the third component[3] of the sample was increased (the system $Pb_xMo_6S_8$) or when another element, say gallium[1,2,5] was added (the system $SnGa_xMo_6S_8$). On the whole such samples were undoubtedly non-single-phase and inhomogeneous, so that it was impossible to establish the necessary connection between the observed values of J_c and the features of the sample microstructure.

We present here the results of investigations of the critical currents of bulky sintered single-phase samples of the compound $PbMo_6S_8$. Of primary interest was the current-carrying capacity of the samples as a function of a microstructure parameter such as the grain size. To compare the results it was necessary to study single-phase samples of as equal a composition as possible, and hence of equal values of the main superconducting parameters.

The single-phase $PbMo_6S_8$ samples for the current measurements were synthesized from powders of Mo, PbS, and MoS_2 at a temperature 900 °C in lead vapor for 60 hours. The single-phase character of such a sample was ensured by choosing the batch composition within the two-phase region of TMS + Pb (see Ref. 6) and was confirmed by x-ray structure and x-ray analysis data. The powder of the synthesized compound was pressed in dismountable matrices at 30 kbar pressure to form cylinders of 5 mm diameter and 12–15 mm height. The samples were then repeatedly annealed in quartz ampoules at 850 °C in the presence of Pb or MoS_2, so that the TSM composition remained unchanged and always corresponded to the vertex of the three-phase region of (TMS + MoS_2 + Pb) on the isothermal tie-line of the Pb–Mo–S phase diagram (see Ref. 6), as confirmed by the constancy of the values of T_c and of $(dB_{c2}/dT)_{T_c}$, namely 13.5 K and 4.35 T/K, respectively. The dependence of the critical current on the magnetic field at 4.2 K was measured after each annealing stage. The critical-current density was measured by an inductive method using a weak trapezoidal modulation of the magnetic field. The experimental setup is described in detail in Refs. 5 and 7. By way of example, Fig. 1 shows the measured $J_c(B)$ at $T = 4.2$ K for one of the samples. It can be seen that such an annealing increases substantially (by 2.5 times) the absolute values of the critical-current density, with practically no change in the character of the $J_c(B)$ dependence. After approximately 200 hours of annealing the current density saturates at the levels 10^9 and 2.4×10^8 A/m^2 in fields 2 and 10 T, respectively.

Investigation of the sample microstructure with a scanning electron microscope has shown that the samples in which annealing produced the maximum critical current were considerably less porous than the initial samples (prior to annealing). It appears that such a heat treatment (annealing at relatively low temperature in the presence of Pb and MoS_2) accelerates the sintering process and consequently makes the samples more monolithic on account of the chemical-potential gradients of lead and sulfur in the gas phase. It should be noted that if the annealing is carried out

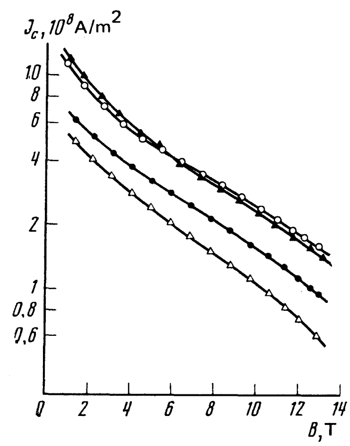

FIG. 1. Dependences of the critical-current density on the magnetic field at 4.2 K for one of the $PbMo_6S_8$ samples after annealings at 850 °C for 48 h: △—first annealing; ●—second annealing; ▲—third annealing; ○—fourth annealing.

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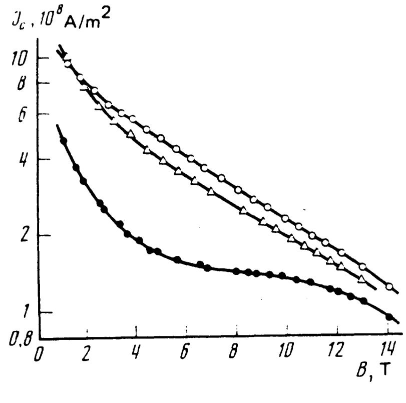

FIG. 2. Dependences of the critical-current density on the magnetic field at $T = 4.2$ K for $PbMo_6S_8$ samples with different grain size: ○—0.3 ± 0.1; △—0.7 ± 0.2; ●—1.3 ± 0.4 μm.

in the absence of Pb and MoS_2 no significant changes in J_c are observed at all (even at an annealing temperature 1150 °C), whereas T_c decreases to 10.7 K, apparently because of noncongruent evaporation of the $PbMo_6S_8$ (Ref. 6).

Once saturation of the critical current was achieved as a result of annealing at 850 °C, samples with different grain sizes were obtained by additional annealing for 24 h in the presence of Pb and MoS_2 at higher temperatures, 1050 and 1250 °C. According to the electron-microscopy, the average grain size d was $0.3 \pm 0.1\,\mu$m in the initial samples annealed at 850 °C, $0.7 \pm 0.2\,\mu$m in samples annealed at 1050 °C, and $1.3 \pm 0.4\,\mu$m in samples annealed at 1250 °C. For such samples with different grain size, the $J_c(B)$ dependence was measured in a wide temperature range and the results in the form of $P_v(b,T)$ plots were analyzed. Thus, Fig. 2 shows the field dependences of $J_c(B)$ at 4.2 K, from which it can be seen that when the grain size is increased the absolute values of J_c decrease and the character of the $J_c(B)$ dependence itself is substantially altered (particularly for samples with $d \approx 1.3$ μm). The dependences of the pinning force on the relative induction are shown in Fig. 3. For initial samples having in our experiments the smallest grain size $d \approx 0.3\,\mu$m, the values of $P_v(b,T)/P_{v\,max}(T) = f(b)$ fit a universal relation $f(b) = b^{1/2}(1-b)^2$ in a wide temperature interval (Fig. 3a). The maximum pinning force $P_{v\,max}(T)$ at the corresponding temperature is in this case the proportional to $B_{c2}^{5/2}(T)$. The scaling law holds also for samples with grain size 0.7 μm (Fig. 3b), but at large b the experimental values of $P_v/P_{v\,max}$ lie somewhat higher than the $f(b) = b^{1/2}(1-b)^2$ curve, and in addition $P_{v\,max}(T) \sim B_{c2}^2(T)$. For the samples with the largest grain size in our experiments, $d \approx 1.3\,\mu$m, the scaling law does not hold and the shape of the $P_v(b)$ curves depend strongly on the temperature (Fig. 3c).

Thus, relatively high values of the critical current can be reached in bulky single-phase $PbMo_6S_8$ samples (at $T = 4.2$ K we have $J_c = 1.6 \times 10^9$ A/m^2 in a field 10 T and 3×10^8 A/m^2 in a field 10 T). It appears that the main pinning centers are here, as in many other type-II superconductors, the grain boundaries. Favoring this assumption is the increase of the current-carrying capacity with decreasing grain size, and the functional form of the $P_v(b,T)$ relations. The anomalous form of the $J_c(B)$ curves and the violation of the scaling laws for samples with large grain size may be due to peculiarities in the behavior of the vortex lattice in the presence of a strongly diluted system of flat pinning centers.

It should be noted in conclusion that the maximum values of J_c were obtained in this study for $PbMo_6S_8$ samples with relatively large grains ($\approx 0.3\,\mu$m). There are therefore definite grounds for assuming that a decrease in the average grain size can lead to further increase of the critical-current density in polycrystalline TMS samples.

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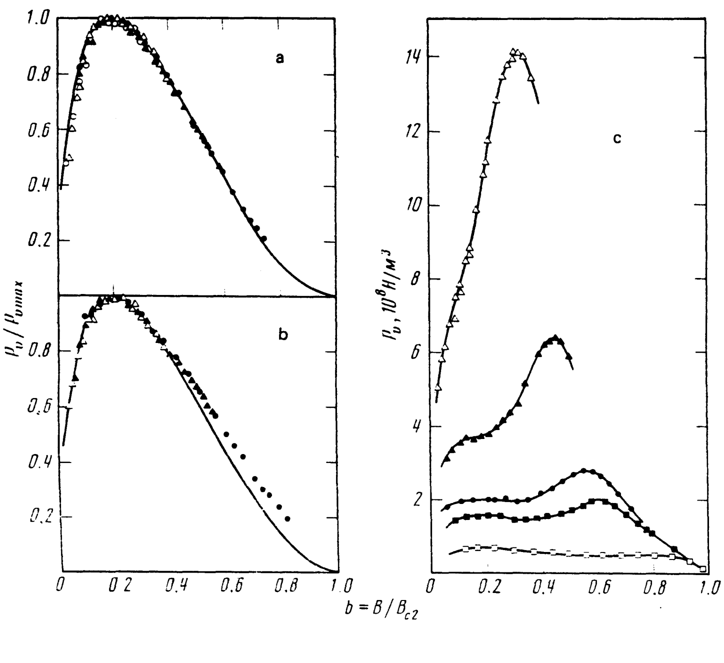

FIG. 3. Dependences of the pinning force on the relative induction at various temperatures: ○—2.4 K; △—4.2 K; ▲—7.0 K; ●—8.5 K; ■—9.0 K; □—10.5 K for samples with grain size 0.3 ± 0.1 (a), 0.7 ± 0.2 (b), and 1.3 ± 0.4 μm (c).

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