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Influence of the constant load sintering on the improvement of the transport critical current in Ag-$(Bi,Pb)_2Sr_2Ca_2Cu_3O_x$ tape form conductor

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Abstract

The effects of the short time, low value constant load pressing process of cold rolled silver clad wires conducted at temperatures close to the partial melting point of the ceramic core, from 800°C to 866°C on the microstructure and critical current anisotropy have been studied in detail.
The strong influence of the constant load sintering at up to 2 MPa for 1 hour on the improvement of the alignment of the ceramic core in the tape form conductors was confirmred by microstructure investigations and also by the I_c versus μ_0H measurements. The magnetic field dependence of the transport critical current, for μ_0H parallel to the surface of the tape and also for μ_0H perpendicular to the surface of the tape, is very significantly reduced in the tapes sintered under constant load. The analysis of the mechanical creep of silver and ceramic core versus temperature allowed us to optimise the densification conditions of the investigated single core Ag-"2223" tape form conductor for the load used .

1. Introduction

The conversion of the $(Bi,Pb)_2Sr_2Ca_2Cu_3O_x$ phase by sintering involves a liquid phase reaction which improves the connectivity between the plate-like grains, but simultaneously it is necessary to repeat the cold rolling and sintering procedure in order to minimise the effect of the retrograde densification which causes volume expansion of up to 10% and a reduction in the density of the superconducting core, reducing also the critical current, I_c, capabilities of the tapes. Because the multi-rolling and multi-pressing procedure is very complex, other more effective techniques should be considered for densification and texturing of the ceramic core in future conductors. The cold uniaxial multi-pressing and subsequent sintering procedure has been proposed previously by Yamada et.al [1] and leads to J_c value equal $2.5*10^4$ Acm^{-2} after three sintering and two cold-pressing procedures at 50 kbar each.

Studies of the densification of the "2223" phase by hot uniaxial pressing of pellets have been conducted by different research procedures : under pressure of 39MPa at 800°C in a vacuum for 2 hours [2] and under pressures 50 and 200 MPa at temperatures from 650°C to 840°C for 2 hours and also under pressure 300 kg cm^{-2} at 822°C for 2h [3]. After additional annealing at 835°C-840°C in air for 40h-100h the achieved J_c value varied from $700Acm^{-2}$ [2] and 1500 Acm^{-2} [4] up to few thousands Acm^{-2} [3] without regular pattern. The hot uniaxial pressing of the screen-printed coatings under pressure $500kGcm^{-2}$ at 855°C for 30 min raise the J_c value to more than 4 kA cm^{-2} [5]. The alternative approach towards hot densification has been presented by use of the Hot Isostatic Pressing of the "2223" compound under 200 MPa pressure of argon gas at 650°C for 2h. The J_c of the HIPed sample was only 1000A cm^{-2} suggesting that HIPing is a technique which may introduce dislocations in the grains and modulated structure, but does not significantly improve the Jc properties of the "2223" phase.

The alternative approach towards densification of the "2223" phase in silver clad wire has been recently presented by Guo et.al [6]. The proposed process consists of a short period of partial melting of the "2223" phase, three sintering-recovery processes at 832°C for a total time of about 200h assisted by three intermediate cold pressing procedures. As a result of that complicated process an increase in the irreversibility line slope has been achieved in comparison with non-melted samples [7]. The above improvement is promising but required multi-sintering and multi-cold pressing procedures which is not very practical for manufacture of long superconducting tapes. In our paper we present investigations of the short time low load ***in-situ*** single stage hot uniaxial pressing and sintering procedure of Ag-"2223" tape in relation to it's transport critical current and microstructure.

2. Experimental

The tape specimen was prepared by the powder in tube technique, successfuly adopted from the previous low

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temperature superconductors. The superconducting powder $Bi_{1.84}Pb_{0.34}Sr_{1.91}Ca_{2.03}Cu_{3.06}O_x$ used in this study was supplied by Merck, UK. The powder was packed into the silver tube OD 6mm with density of 4.2gcm^{-3} (64% of the theoretical density of the "2223" phase) and swaged to the diameter 1.75mm and finally cold rolled to the tape form with thickness of 0.25mm under controllable conditions. The whole hot uniaxial pressing and sintering process has been conducted over a temperature range from 800°C to 866°C for 9h and 50h, in a vertical furnace in air. Three different times of HUP have been chosen: 5min, 15min and 60 min. The value of the induced pressure was 2MPa. Prior to the HUP the surface of the silver clad tapes was coated with a MgO layer which not only protects the tape against bonding to the metal-ceramic die but assures that distribution of the force over the length of the tape was uniform.

The degree of alignment of the ceramic core has been defined after peeling off the silver cover (for the ceramic layer peeled off with silver and and the opposing surface of the remaining ceramic core) by comparison of the I(119)/I(00$\underline{14}$) XRD peak intensities for the ceramic core after different stages of densification. The partial melting temperature was defined by DTA measurements under a heating rate of 10°Cmin^{-1}. The changes in density of the ceramic core after sintering-HUP procedure were evaluated using an Image Analysis System, on the basis of the changes in cross section of the internal hole of the silver sheet in the final tape after cold rolling where density was 66.5%. The density of the superconductor core was calculated from $\rho_s=(W_o-W_{Ag})/A_{s,ave}\ l$, where $A_{s,ave}=A_o-(W_{Ag}/\rho_{Ag}\ l)$, W_o is the total weight of the tape, l is the length of the tape, W_{Ag} and ρ_{Ag} are the weight and density of the silver respectively (obtained after dissolving the superconductor core), A_o is the owerall cross-sectional area of the silver clad tape as measured with Image Analysis System. The microstructure and chemical composition of the ceramic core were examined by SEM/EDX. The recovery process of the "2223" phase at different stages of sintering was investigated by the AC susceptibility technique for magnetic field μ_0H=1Gauss. The I_c measurements versus different orientation of the external magnetic field to tape surface have been conducted by a fully computerized unit, with accuracy E=0.01μVcm^{-1} for all investigated tapes.

3. Results and discussion

The change of density of the ceramic core after sintering-HUP-sintering process for 9h and only sintering process for 50h at different temperatures is presented in Figure 1. It becomes apparent that sintering of the samples without load does not cause densification of the ceramic core.

The DTA measurements of the initial powder and silver clad wire after the final cold rolling prove that silver causes lowering of the melting point of the "2223" phase, Figure 2, which is in good agreement with data presented by Oota et.al [8]. According to our previous investigations the processes at the silver-"2223" interface are responsible for lowering the melting point of the "2223" phase [9]. The optimum temperature for solid state sintering of the "2223" phase in silver clad wire is around 828°C, Figure 2, but in the case of the procedure used, partial melting of the "2223" ceramic core at 853°C (Figure 2) reinforces the creep of the conductor under HUP.

The cross sections of the samples sintered alone at 850°C for 50h and the sample sintered + HUP at 850°C for 9h are presented in Figure 3.

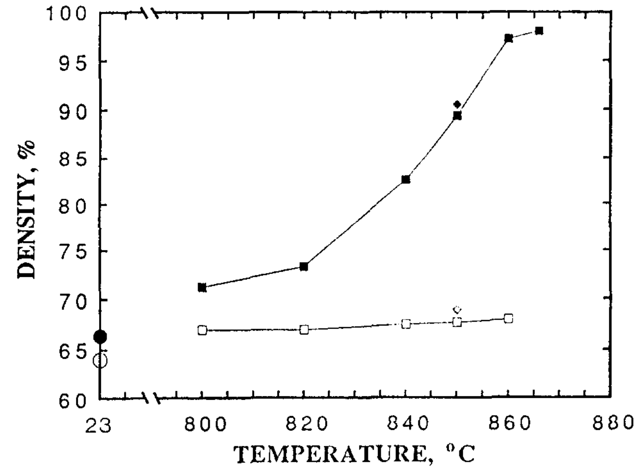

Figure 1. Density of the ceramic core versus processing temperature for the Ag-"2223" tapes ; ○ after packing in the silver tube,● after final cold deformation, □ sintering (9h), ■ sintering (8h) + HUP (1h), ◇ sintering (50h), ◆sintering (8h) + HUP (1h) + sintering (41h) .

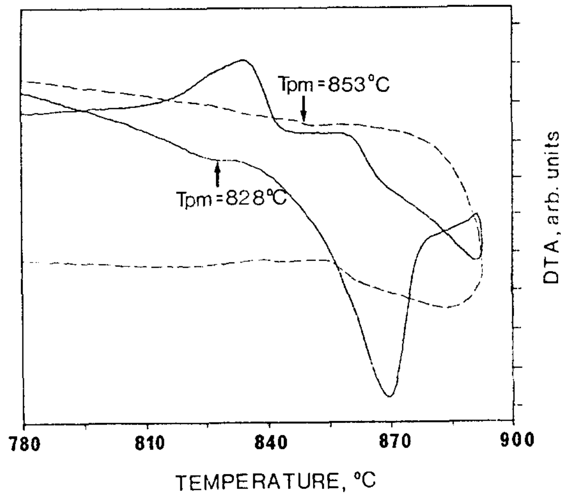

Figure 2. DTA thermograms of: — —superconducting "2223" powder in Al_2O_3 crucible, — silver clad "2223" tape after final cold rolling.

The pronounced reduction of the whole tape thickness after HUP is a consequence of creep of the ceramic core. The creep of the conductor at high temperature under uniaxial load causes pronounced densification of the ceramic core and improvement of alignment of the "2223" plates.

Without load the silver sheet does not undergo detectable creep and consequently the dimensions of the internal hole in conductor remain unchanged.

Microprobe analysis of the ceramic at different stages of the wire preparation indicates that the impurity phases which coexist with "2223" phase are present in all wires independently of the preparation procedure, Figure 4, however the distribution of impurities differs for the HUP samples suggesting that dispersion-coagulation-reformation processes of such phases may take place. The above effect requires further study in respect to the role of the impurity distribution on the pinning of flux lines in the ceramic core and grain boundary composition.

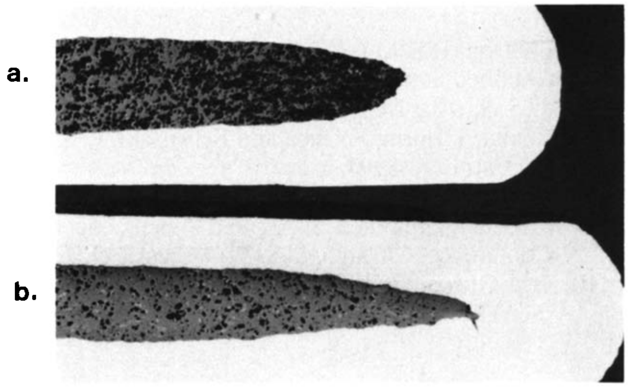

Figure 3. The cross-section of the Ag-"2223" tape; a/ sintering (50h), b/ sintering (8h) + HUP (1h)

The degree of alignment of the ceramic core at different stages of the tape manufacture is presented in Table 1.

Table 1. Degree of alignment in the "2223" ceramic core

	Degree of alignment $I(00\underline{14})/I(119)$	
Sample	Ag-"2223" interface	central "2223"core
"2223" powder	0.54	
rolling	1.8	2.9
50h sintering	22	8.4
sintering (8h) +HUP (1h) + sintering (41h)	24	24

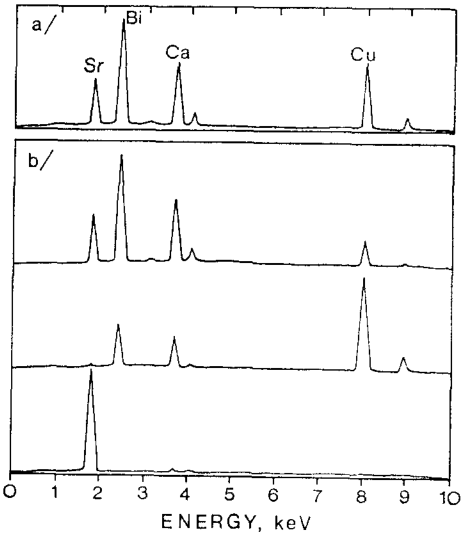

Figure 4. EDX analysis of the ceramic core ; a/ "2223" matrix, b/ impurity phases.

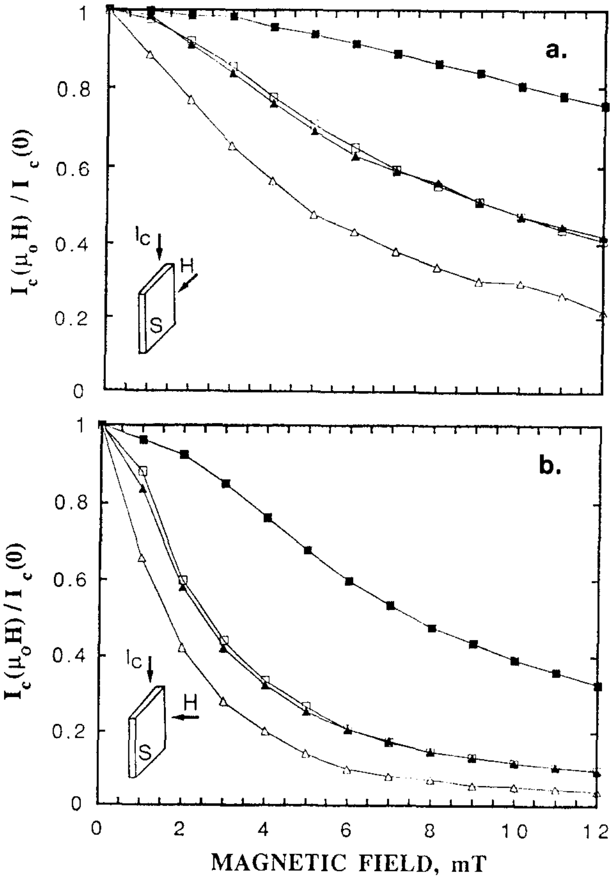

Figure 5. Plot of the reduced critical current versus magnetic field for Ag-"2223" tapes prepared by different hot processes; a/$I_c \| \mu_oH$, b/$I_c \perp \mu_oH$; △ sintering (9h) {Jc=0.54 kAcm^{-2}}, ▲ sintering (8h) + HUP (1h) {J_c=1.7 kAcm^{-2}}, □ sintering (50h) {Jc=2.3kAcm^{-2}},
■ sintering (8h) + HUP (1h) + sintering (41h) {J_c=6.5kAcm^{-2}}

The hot pressed ceramic presents the highest degree of alignment along the a-b plane and also it became apparent that the ceramic layer close to the silver-"2223" interface is better aligned than the cental crramic core, which confirms the previously reported influence of the Ag-"2223" interface densification processes [9,10].

The critical current density of the ceramic core, only sintered for 50h at 850°C was 2.3kAcm^{-2} but J_c value of the in-situ sintered-HUP-sintered "2223" core was 6.5kAcm^{-2}. Strong improvement of the I_c versus μ_0H characteristics for the HUP samples, Figure 5, suggests improvement of the grain alignment in the ceramic core and some improvement in connectivity between the grains.

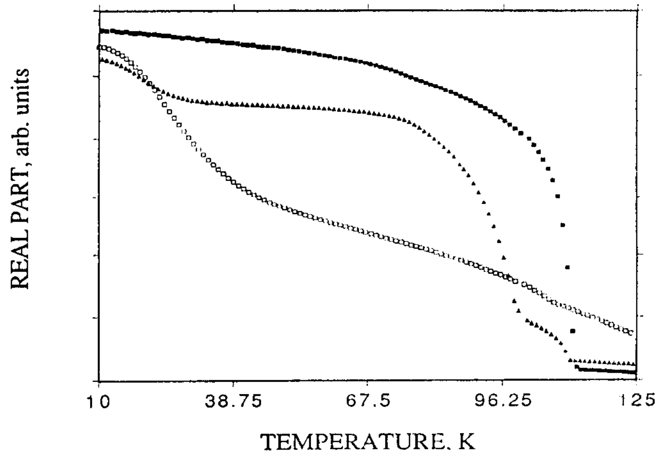

Figure 6. Superconducting diamagnetic AC susceptibility of the "2223" phase: ■ starting powder, □ after final cold reformation, ▲ Ag-"2223" tape after sintering (8h) + HUP (1h)+ sintering (41h); temperature 850°C

The comparison of the AC susceptibility of the sample, after ***in-situ*** hot uniaxial processing, and the initial superconducting powder indicates that the final sintering process of the "2223" phase after HUP required further optimisation, Figure 6.

4. Conclusions

The process route presented here, of ***in-situ*** sintering - Hot Uniaxial Pressing - sintering appears to be very promising in respect of improvement of the density of "2223" phase and also increase of J_c value and the reduction of J_c vs μ_0H dependence.

The achieved results suggest that in future superconducting reinforced conductors, compression force should be induced during sintering to improve intergrain connectivity and volume densification of the ceramic core if only single-stage sintering has to be used, eliminating that way the multi-rolling and multi-sintering procedure.

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