CHAPTER V

INFILTERATION GROWTH PROCESS FOR Sm-123

5.1. INTRODUCTION:

Melt processing of REBa$_2$Cu$_3$O$_y$ (123) system results in improved microstructure and current density ($J_c$) [1-3]. Inspite of these advantages, the melt processed samples however have some inherent defects like shrinkage and macrodefects such as cracks, voids and pores, etc. [4]. Those defects limit the $J_c$, and also limit making as devices for practical applications. In melt processing, the RE-123 phase is decomposed above its peritectic decomposition temperature, where it incongruently melts into a solid RE$_2$BaCuO$_5$ (211) and liquid phases. The surface tension forces exerted by the newly formed liquid shrinks the sample ~ 20-25 %. Moreover, pores and voids form, because of the liquid phase outflow. This melting is accompanied by a change in the dimensions of the sample, which is upto 25% [5]; distortions and cracks may also appear along with the shrinkage.

In ceramic processing techniques like directed metal oxidation (DIMOX) and pressure-less metal infiltration (PRIMEX) [6,7], the molten liquid alloy is allowed to infiltrate or percolate into a semi-porous preform. The advantage of these processing techniques is that large near-net shaped components can be produced, which are free of macro-defects like shrinkage, porosity, etc. The samples need minimal machining after the growth process. On similar lines, recently, a novel processing route has been developed known as the Infiltration-Growth (IG) process [5,8]. In the IG process, the possibility of making near-net shaped high $J_c$ components of Y-123 and Gd-123 superconductors has been
demonstrated. In the IG process, a preform of $\text{RE}_2\text{BaCuO}_3$ (RE = Rare earth) is kept in contact with a source of liquid phases and melt processed. In this chapter the extension of that work to the Sm-123 system is discussed. The Sm-123 system needs to be processed in oxygen-poor atmosphere to reduce the formation of low-$T_c$ solid-solution phases. By processing in reduced oxygen atmosphere the solid-solution formation can be suppressed and the resulting samples will have $T_c$ above 90 K, with high current density [9].

5.2 EXPERIMENTAL DETAILS

The precursor powders were prepared by solid-state sintering route. The Sm-211 and Sm-123 powders were prepared by taking required amounts of $\text{Sm}_2\text{O}_3$, $\text{BaCO}_3$ & CuO and mixing them thoroughly in a centrifuge mill using an organic liquid as the medium. The dried powders were sintered at 975°C and 930°C to form Sm-211 and Sm-123, respectively. The sintering was repeated twice to achieve phase purity. Two different configurations were tried for the IG process. In one configuration the Sm-211 preform was kept below the 123 pellet, in this configuration the liquid will infiltrate into the 211 preform at the high temperature melting and the usual growth process of 123 takes place. In a second configuration the 211 preform was kept above the 123 pellet and the usual melt processing was done. In the second process named as IG-2, the liquid will percolate into the 211 preform based on the capillary action and the growth of 123 will take place. These were melt processed by melting at 1115°C for 10 minutes, and then slow cooled from 1060°C to 950°C at a rate of 1°C/hr, all under flowing Argon gas. The
samples were oxygenated in flowing oxygen atmosphere according to the schedule described in the experimental chapter.

5.3. PRELIMINARY CHARACTERISATION

The X-ray diffraction patterns (Fig. 5.1) of the two samples showed the formation of the Sm-123 phase with some amount of Sm-211. The lattice parameter values matched that reported for Sm-123 values [10], indicating that the solid-solution formation has been suppressed to a large extent. The $T_c$ (Fig. 5.2) of the samples were above 90 K, confirming that the solid-solution formation had been suppressed to a large extent. During the melt processing the Sm-123 melted and the liquid infiltrated the Sm-211 preform. Slow cooling through $T_p$ produced void-free specimens (as seen from cutting the samples), with large domains of 123 visible to the naked eye. The dimensions of the samples of the samples showed no change after melt processing.

5.4. MICROSTRUCTURAL FEATURES

Samples were quenched just after melting at 115°C to study the infiltration of the liquid phases into the Sm-211 preform. Fig. 5.3 shows the typical microstructure of a quenched sample using an SEM. The particles appearing white are the Sm-211 phase, and the black background is the liquid phase. The SEM image shows that the liquid phases completely fill the vacant regions between the blocky Sm-211 particles. The ease with which complete infiltration has occurred without the application of an additional driving force like pressure encourages from the view point of fabricating large samples with the 1G process. The particle size of
Fig. 5.1: XRD patterns of the IG processed samples showing the presence of Sm-123 and Sm-211 phases.
Fig. 5.2(a): Resistivity vs temperature plot of the IG processed sample. The $T_c$ of the sample is 90.4 K.

Fig. 5.2(b): Resistivity vs temperature plot of the IG-2 processed sample. The $T_c$ of the sample is 90.1 K.
Fig. 5.3 : SEM image of the sample quenched after the infiltration of liquid phases into the Sm-211 preform.

Fig. 5.4 : Optical micrograph of the IG processed sample indicating less porosity in the sample.
the Sm-211 in the liquid is very small and there has not been a large coarsening of such particles in the liquid. Fig. 5.4 shows the optical micrograph of the as polished melt processed sample. The micrograph shows a uniform distribution of the Sm-211 particles. The uniformity of distribution and the possibility of tailoring the 211 size is an advantage with the process. Also, the sample has less porosity and voids, due to the reasons mentioned above. Fig. 5.5 is an SEM image at higher magnification. The average size of the Sm-211s in the textured samples is approximately 1 μm. Fig. 5.6 is a fractured surface of the sample, which shows parallel Sm-123 grains in the sample. Fig. 5.7 is the SEM image of the sample prepared by a second configuration (1G-2) in which the Sm-211 preform is kept above the source of liquid phases. The liquid phases move up into the Sm-211 preform by the capillary action. Similar features are seen in this sample as seen in Fig. 5.5. From the SEM images the volume percentage of the 211 particles occupied in the microstructure is evaluated using standard metallographic techniques [11]. The volume percentage is ~ 70 % in both the samples, this corresponds to a sample with starting composition of Sm-123 with 40 mol % Sm-211 for melt processing. This conclusion is arrived on the basis of the data from the micrographs of melt processed Sm-123 with increasing concentrations of Sm-211 (0 to 40 mol % Sm-211, chapter III), where the features like platelet width and gap width match that of the 1G processed samples. However, the difference in the 1G sample is that the 211 particles are highly refined in the microstructure, where as, in conventionally melt processed sample of Sm-123 with 40 mol % of Sm-211, the 211 particles are much larger in size.
Fig. 5.5 : SEM image of the IG processed sample showing the usual features of melt processed samples.

Fig. 5.6 : SEM image of the fractured surface of IG processed sample showing the alignment of the grains.
5.5. CURRENT DENSITY

The dc magnetic hysteresis (M-H) loops recorded at 5 K, 35 K & 77 K, for field applied normal to the sample surface are shown in Fig. 5.8. No anomalous features like peak-effect were present, as observed in the stiochiometric RE-123 melt processed samples [13,14].
Fig. 5.8(a).: M-H loops of the IG processed samples at 5 K.

Fig. 5.8(b).: M-H loops of the IG processed samples at 35 K.
Fig. 5.8(c). M-H loops of the IG processed samples at 77 K.
The $J_c$ is evaluated using the Bean's critical state model from the loops recorded at various temperatures, is plotted in Fig. 5.9, against the field. The flux pinning force was calculated using the relation, $F_p = J_c B/10$. The data at 77 K are plotted in Fig. 5.10, which clearly shows that the IG sample has a higher pinning force. The variation of the normalised pinning force ($F_p/F_{p_{\text{max}}}$) with the normalised field $b$ ($B/B^*, \ B^*\text{= max. field}$) is plotted in Fig. 5.11. The two samples have a peak around 0.2 [15].

Fig. 5.9(a). $J_c$ vs B plots of the samples at 5 K.
Fig. 5.9(b). $J_c$ vs B plots of the samples at 35 K.

Fig. S.9(c). $J_c$ vs B plots of the samples at 77 K.
Fig. 5.10: Plot of the variation of the pinning force with field at 77 K.
Fig. 5.11: Plot of the variation of the normalized pinning force with normalized field at 77 K.

5.6. IRREVERSIBILITY LINE

The irreversibility line is for the IG processed sample is plotted in Fig. 5.12. The IR line is less steeper in comparison to the samples showing high $J_c$ in the Sm-123 system, indicating that the pinning strength of the sample is low. This can also be shown from Fig. 5.10. The IR line has a slope change at a field around 0.4 Tesla.

5.7. MAGNETIC RELAXATION

The magnetic relaxation rate of the sample is measured at a lower temperature. Fig. 5.13 shows the variation of the normalized relaxation rate with field at 30 K. The relaxation rate initially decreases upto a field of 0.3 Tesla and later increases with field, but the relaxation rate has a slope change at a field of 2.8
In Fig. 5.14, the $J_c$, flux pinning force and the relaxation rate are plotted together against the field. The $J_c$ decreases and the relaxation rate increases with field. The flux pinning force has a peak in the 2-3 Tesla region. It is known that, the behaviour of the $J_c$ in superconductors is dominated by pinning in the low fields and by the relaxation rate in high fields [16]. In the IG processed sample such similar

Fig. 5.12 : Irreversibility line of the IG processed sample for H parallel to c-axis.
Fig. 5.13 : Variation of the relaxation rate with applied field at 30 K for the IG processed sample.

Fig. 5.14 : Comparative plot $J_c$, flux pinning force ($F_p$) and relaxation rate ($S$) against the applied field at 30 K.
effect is observed. The creep rate has a slope change at a field above the point at which the flux pinning force has a peak. This indicates the pinning force saturates at the peak value, and above that field, relaxation rate will dominate the properties.

5.8. CONCLUSIONS

The aim of this work was to demonstrate that the IG process is a viable way of melt processing Sm-123 phase with good microstructural features. There is an improvement in the refinement of Sm-211 inclusions, but the $J_c$ is not very high. It is proposed that the low $J_c$ in the IG processed Sm-123 sample is primarily due to the large volume percentage of Sm-211 in the microstructure. The $J_c$ dependence in the Sm-123 melt processed samples on increasing Sm-211 content has been found to have two peaks (chapter III, Fig. 3.14) \[12\]. A major peak exists at the 10 mol % Sm-211 concentration, and a minor peak at 30 mol %. As discussed in chapter III, low at concentrations of 211, an additional field induced pinning from the low-$T_c$ oxygen deficient regions helps to enhance the $J_c$. For increasing 211 content (above 20 mol %), the field induced pinning is absent, and the $J_c$ decreases \[14\]. This is unlike in the Y-123 and Gd-123 \[17,18\] systems where the maximum in $J_c$ as a function of 211 concentration occurs at - 30 mol % of 211. In these systems the predominate contribution to the pinning comes form the defects at the 123/211 interface.

It is proposed that, in the present IG samples the relatively high concentration of Sm-211 has suppressed the spinodal transformation (chapter III) and the pinning in it is predominantly from the 123/211 interface defects. The pinning from such interfaces does not contribute to $J_c$ enhancement substantially.
in the LRE-123 systems as is evident from the present experiments (chapter III) and the work of others [19].

Because of the dramatic advantages of possible shape forming in combination with $J_c$ enhancement by the IG process, it is highly desirable to enhance the $J_c$ further in IG processed Sm$\cdot$123. A possible way of doing as suggested by the present work is to reduce the Sm$\cdot$211 content in the microstructure.
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