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Structural investigations in 2:17 type SmCo alloys

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Abstract: Structural transformations at various stages of thermal processing in powder metallurgically (PM) processed 2:17 type SmCo alloys have been investigated by X-ray diffraction (XRD), optical, scanning and transmission electron microscopes (SEM and TEM). It is observed that 2:17 phase with hexagonal $TbCu_7$ (1:7H) + partially transformed rhombohedral Th_2Zn_{17} (2:17R) structures exist at temperatures ≥ 1463 K (at Sm ≥ 10.5 at %). It has also been observed that in Sm rich compositions, many grains of 2:17 phase having 1:7H structure exhibit lamellar feature after solution treatment. Such microstructures result in good combination of permanent magnet properties, after isothermal aging and slow cooling treatment.

Key words: *2:17 type SmCo, coercivity, microstructure, domain wall pinning*

1. Introduction

High performance permanent magnets of 2:17 type SmCo with additions of Cu, Fe and Zr are complex metallurgical system and are developed by powder metallurgical (PM) technique with stringent control on alloy chemistry and thermal process parameters [1]. The hard magnetic properties in these alloys are attributed to the development of a fine scale cellular precipitate microstructure achieved through a multi-stage heat-treatment procedure, *viz* sintering and solution treatment (1433 – 1483K) and isothermal aging (1073 – 1148K) followed by slow cooling to 673K. The fine scale microstructure consists of rhombohedral $Sm_2(CoFe)_{17}$ [2:17 R] phase as cells surrounded by hexagonal $Sm(CoCu)_5$ [1:5H] boundary phase with Zr-rich platelet phase running across many cells and cell boundaries [2]-[5]. The cell boundary and the platelet phases act as pinning centres for magnetic domain walls and the material exhibits permanent magnet characteristics. The exact mechanism of cellular precipitation and the structure dependency of coercivity are still the topics of research. It has been reported that in 2:17 type magnets, formation of the $TbCu_7$ (1:7H) structure at the solution treatment stage is essential for the development of large intrinsic coercivity ($_iH_c$) and is due to larger anisotropy of the 1:7H structure than that of Th_2Zn_{17} (2:17R) type structure [6]. As the formation of 1:7H structure is sensitive to temperature and Sm content in the alloy, it is of scientific interest and of technological importance to study the phase formation in 2:17 SmCo system at various stages of heat-treatment and its dependence on magnetic properties. In the present study, XRD and microstructural investigations carried out on phase transformations in 2:17 SmCo alloys are reported. The structural data obtained are correlated to the magnetic properties of the alloys.

2. Experimental

Samples of 2:17 type SmCo magnets with nominal compositions of Co-(9-12)Sm-(18-21)Fe-(4-5)Cu-(2-4)Zr (in at%) were prepared by PM technique coupled with magnetic field alignment as reported elsewhere [7]. The green compacts were sintered and solution treated at 1448 – 1483K and subsequently subjected to isothermal aging at 1123 K for 10 h and slow cooled to 673 K at the rate of 30 K/h. X-ray diffraction (XRD)patterns were obtained using a Philip X-ray powder diffractometer, PW3020 with CuKα radiation. Microstructural studies were carried out using optical, scanning electron (Leo 440i SEM) and transmission electron microscopes (Philip EM 430T operating at 200 kV). Magnetic measurements were performed on disk samples using auto hysteresis graph (Walker Scientific Inc.).

3. Results And Discussions

A large number of 2:17 alloys of different compositions in the range as mentioned in section 2, have been processed and detailed structure-property correlation has been carried out on each alloy system. It has been observed that alloys with Sm-rich (> 10.5 at%) compositions only could yield good permanent magnetic properties with large $_iH_c$ and therefore in the present study, structural and magnetic

investigations on alloy with composition of 11Sm-60.5Co-21Fe-4.5Cu-3Zr are reported.

3.1 XRD studies

The three crystal structure types of 2:17 phase, *viz.* (i) hexagonal Th_2Ni_{17} (2:17H), (ii) rhombohedral Th_2Zn_{17} (2:17R) and (iii) hexagonal $TbCu_7$ (1:7H) are modifications of the structure of $CaCu_5$ type [8] and their presence can be easily evidenced from XRD patterns. Th_2Ni_{17} and Th_2Zn_{17} type structures are identified with the presence of (203) and (204) reflections respectively and the absence of these reflections is a finger print for $TbCu_7$ type structure.

Fig. 1 shows XRD patterns of the selected 2:17 alloy quenched from sintering and solution treatment temperature at 1448 – 1483K. It can be seen from the absence of

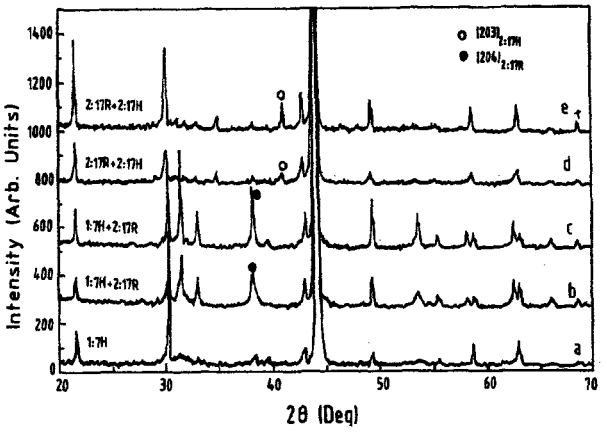

Fig. 1XRD patterns of PM processed 2:17 samples in sintered and solution treated conditions. (a) 1483 K (b) 1473K (c) 1463K (d) 1458K and (e) 1448K. XRD pattern reveals that the samples after solution treatment at ≥ 1463K have a mixture of 1:7H + 2:17R structures.

$(203)_{2:17H}$ and the presence of $(204)_{2:17R}$ reflections [Fig. 1(a-c)] that the samples solution treated in the temperature range of 1463 – 1483K have either 1:7H or a mixture of 1:7H + partially transformed 2:17R structures. In the case of samples solution treated at temperatures < 1463K, the XRD patterns [Fig. 1(d and e)] revealed a mixture of 2:17H and partially transformed 2:17R structures. An attempt has been made to quantify the fraction of 2:17R and 2:17H phases present by estimating the ratio of integrated intensity of $(204)_{2:17R}$ to $(203)_{2:17H}$ reflections at different solution treatment temperatures and the variation is presented in Fig. 2a. It can been seen that the intensity ratio peaks at 1463K, suggesting that for 2:17 samples, solution treatment at 1463K yields maximum volume fraction of 1:7H or 1:7H + 2:17R. On the other hand, in the case of 2:17 alloys where Sm content is < 10.5 at%, the samples after solution treatment showed only 2:17H type structure (not presented).

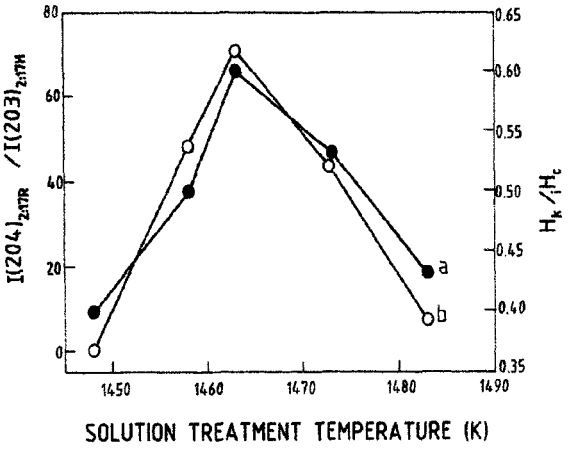

Fig. 2 Plots of (a) XRD intensity ratio of $(204)_{2:17R}$ to $(203)_{2:17H}$ and (b) H_k / $_iH_c$ as a function of solution treatment temperature. It can be seen that both the ratios peak at 1463K

Such samples, upon heat-treatment have not exhibited appreciable magnetic properties. These structural transformations can be understood through the pseudo binary phase diagram (Fig. 3) reported by Morita et al.[8]. As seen from Fig.3, the

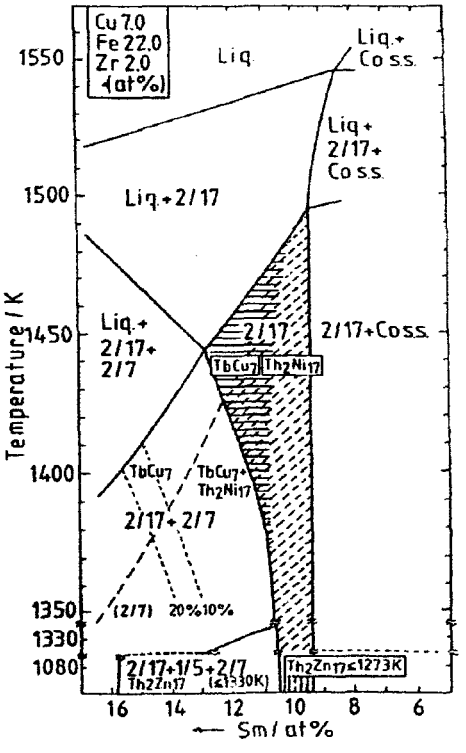

Fig. 3 Vertical section of the quinary Sm-Co-Fe-Cu-Zr phase diagram [8]

2:17H to disordered 1:7H or [1:7H + partially transformed 2:17R] structure depending on the cooling rate. Subsequent isothermal aging at 1123K will result in the transformation of ordered 2:17R structure.

3.2 Microstructural investigations

The evolution of microstructure at various stages of thermal processing (as-cast, sintering, solution treatment, isothermal aging and cooling) is shown in Fig. 4. The as-cast structure (Fig. 4a) consists of three phases: 2:17 matrix phase, a Sm-rich (1:5 type) boundary phase and a Zr-rich needle like phase embedded in the boundary phase. The microstructure (Fig. 4b) of sintered samples showed nearly single phase 2:17 grains. It is interesting to observe that the samples after solution treatment at 1463K

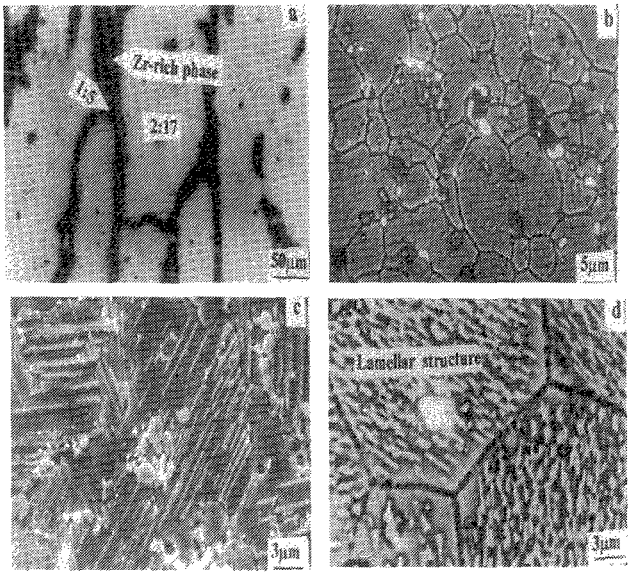

Fig. 4 Microstructures of 11Sm-60.5Co-21Fe-4.5Cu-3Zr alloy (a) as-cast structure (b) sintered at 1483K (c) solution treated at 1463K and (d) isothermally aged at 1123K and slow cooled to 673K. Presence of lamellar feature is seen in many of the 2:17 grains in isothermally aged samples.

reveal lamellar feature (Fig. 4c) in many of the 2:17 grains and subsequently after isothermal aging, they exhibit dense lamellae (Fig. 4d). In the case of samples solution treated at temperatures < 1463K and also in alloys with Sm content < 10.5 at%, such lamellar structure was not observed. The formation of lamellar microstructure could be a result of partial transformation of 2:17R structure from high temperature 1:7H phase. XRD studies on samples at various solution treatment temperatures also indicate a similar structural transformation. TEM studies carried out on isothermally aged sample of selected composition revealed cellular precipitate microstructure with Zr-rich long platelets running across many cells and cell boundaries (Fig. 5). Electron diffraction pattern obtained from the platelet phase revealed that it is of 1:7H type structure.

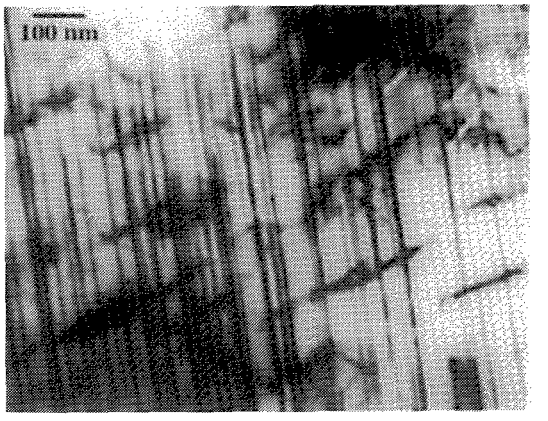

Fig. 5 TEM micrograph showing cellular precipitate microstructure with thin platelets of Zr-rich phase running across many cells and cell boundaries.

3.3. Magnetic characterisation

The samples after isothermal aging and slow cooling (solution treated at different temperatures) have been magnetised to saturation in a pulse magnetic field of 6400 kA/m and the second quadrant characteristics are plotted. While all the samples exhibited an intrinsic coercivity of > 1300 kA/m, a systematic variation in knee field (H_k) has been observed. As $H_k/_iH_c$ represents the squareness ratio of the demagnetisation curve, an attempt has been made to plot the squareness ratio with solution treatment temperature. The $H_k/_iH_c$ behaviour as function of solution treatment temperature is plotted in Fig. 2b. It can be seen that $H_k/_iH_c$ ratio also peaks at 1463K. This indicates that the domain wall pinning is effective when many of the 2:17 grains exhibit lamellae feature.

4. Conclusion

From the structural investigations carried out on powder metallurgically processed 2:17 SmCo alloys, the following conclusions can be drawn.

(i) Large intrinsic coercivities in 2:17 type SmCo magnets are realised only in Sm-rich compositions (Sm ≥ 10.5 at%)

(ii) It appears that there exists a close structure-property correlation between the three aspects *viz.* (a) the necessity of 1:7H or 1:7H + partially transformed 2:17R structures at the solution treated stage, (b) the lamellar feature after isothermal aging and (c) the squareness ratio of the demagnetisation curve.

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