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A Study of Sintering Temperatures Variation on Microstructure Development of Strontium Hexaferrite Millscale-Derived

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Abstract: This paper describes the effect of controlling sintering temperatures on microstructure development of strontium hexaferrite from millscale derived. The sample was prepared by the recyling the waste steel-making product from Malaysian steel factories. Using a Curie temperature separation technique, the wustite, FeO contained in the millscale was separated by Curie temperature separation technique using deionized water at 90°C/100°C in the presence of 1T external field. The wustite was then oxidized in air at 400°C/500°C/600°C for 10 hours. An XRD phase analysis showed that a very high percentage of Fe_2O_3 was present in the final powder preparation. A conventional processing method was then done to prepare hexagonal $SrFe_{12}O_{19}$ pallet shaped samples. The samples were sintered at different temperatures (1200°C, 1250°C, 1300°C and 1350°C) in air for 10 hours. The effect of controlling sintering temperature on orientation grain growth (0.3μm-0.7μm) in $SrFe_{12}O_{19}$ was confirmed by the microstructure surface scan using SEM and was in good agreement with the XRD analysis based on the peak intensity of the (107) plane. Analysis of samples was also done on grain size, Curie temperature and density.

Key Words: Sintering Temperatures, Microstructure, Strontium Hexaferrite

Introduction

Strontium hexaferrite ($SrFe_{12}O_{19}$-magnetoplumbite structure) is a hard magnetic materials have fairly large magnetocrystalline anisotropy, high Curie temperature, large saturation magnetization and a good chemical stability (Babu and Padaikathan, 2001). It has been extensively used as permanent magnets for application of microwave devices in millimeter waveband, in addition to their wide applications and large amount production as permanent magnet and as high frequency core materials (Zhai *et al.*,1989). One of the important properties of hexaferrites is the magnetic properties which is very much dependent on the processing route and microstructure grain growth development (Babu and Padaikathan, 2001; Zhai *et al.*, 1989).

Sintering is the most important factor for the required crystal structure, oxidation state, micro structure and physical condition of the sample. Sintering cycles is important for some applications; there are:

1: Complete formation of the lattice,
2: attainment the chemical homogeneity and
3: attainment of uniform microstructure.

The other properties that extremely depend on sintering temperatures are

1: Average grain size and distribution
2: porosity and locations of pores (inter or intragranular)
3: Density
4: Metal ion valency and
5: Grain boundary conditions (chemistry and thickness of the boundary) (Ghate and Goldman, 1990).

The microstructure control on hard ferrite during fabrication process is a critical desirable properties. These factors can be grouped roughly into two categories. One is the processing condition and the other is the intrinsic property of raw materials. Processing conditions include temperatures, pressure, atmosphere and time. The intrinsic properties are the characteristics of raw powders such as powder purity, powder size distribution, shape distribution, surface area and etc. Powder size is the most dominant factors affecting intrinsic properties (Knudsen, 1959). This premise work however deals with the effect of sintering temperature on the microstructure of the $SrFe_{12}O_{19}$ samples.

Experimental Procedure: The millscale was crushed for 22 hours to obtain the precise sized of powder. Two types of separation was done; the Magnetic Separation Technique (MST) and the Curie Separation Technique (CTST). In the first technique, the crushed millscale powder was separated to magnetic and non-magnetic particles. The crushed powder was placed in the glass tube filled with deionized water in the presence of 1 Tesla external field. The non-magnetic particles as well as all the impurities would drop at the bottom of the tube. The magnetic slurry (MST) was then again poured in the glass tube filled with 90°C- 100°C of deionized water. The weak susceptibility of ferromagnetic particles, assuming to be FeO (wustite), will drop to the bottom of the tube. The FeO was oxidized at 400°C/500°C/600°C for 10 hours in air. The yield of oxidation, Fe_2O_3, was used as a raw material in $SrFe_{12}O_{19}$ pallet fabrication. The $SrFe_{12}O_{19}$ specimens were prepared from powder reagent $SrCO_3$ and Fe_2O_3. The approximation amount of powders were mixed and milled for 20 hours. The mixtures were calcined at 1200°C for 10 hours, reground to fine powders and cold pressed into pallet shape of 1cm diameter and 1cm thickness at ≈70MPa pressure. The pallets were then sintered at 1200°C, 1250°C and 1300°C in air for 10 hours. The surface microstructure scan was studied using Scanning Electron Microscopy (SEM).

X-Ray Diffraction (XRD) was also performed to confirm the Fe_2O_3 phase. Density measurement was carried out

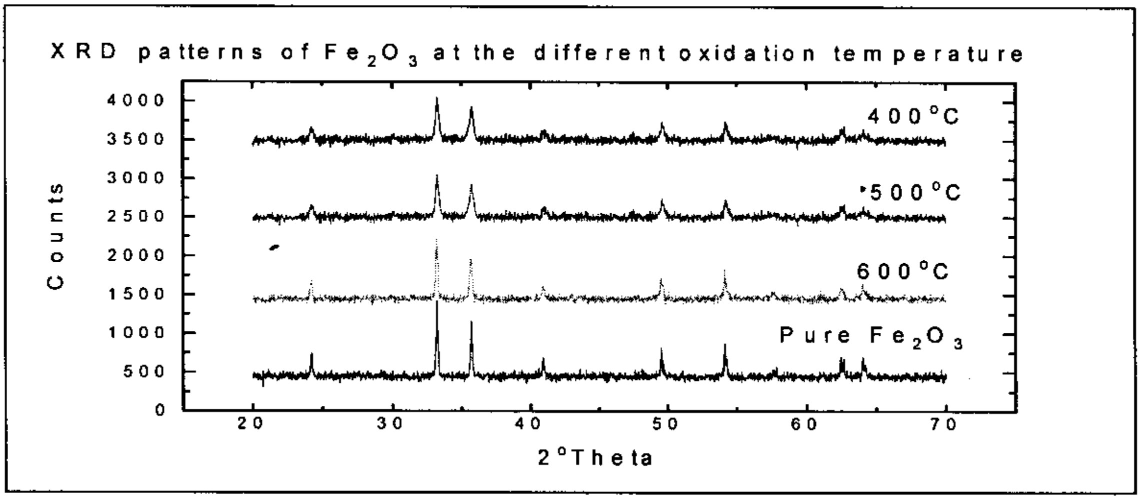
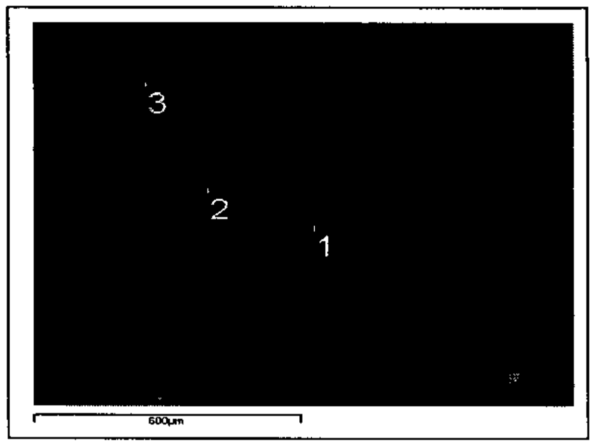
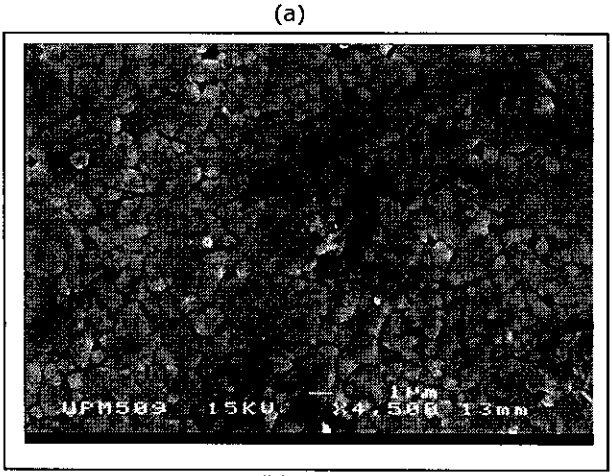


Fig.1: XRD Phase for Fe_2O_3 Derived from Millscale and Pure Fe_2O_3 Powder

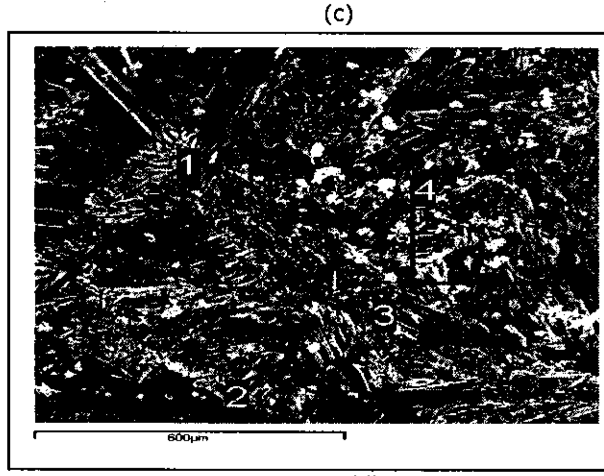
(a)



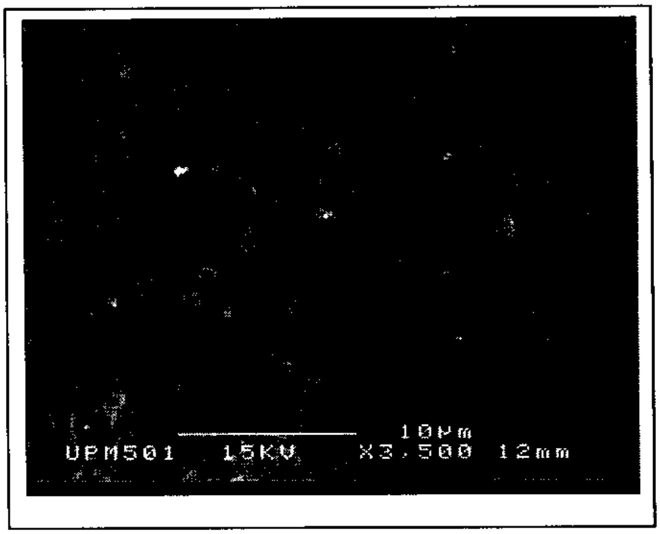
(c)



(b)



(d)



(e)

Fig.2: Scanning Electron Micrograph on Sample Sintered at Different Sintering Temperature, (a) SFM1200 (b) SFM1250 (c) SFM1300 and (d) SFM1350 (e) SFP1200 (High Purity Fe_2O_3)

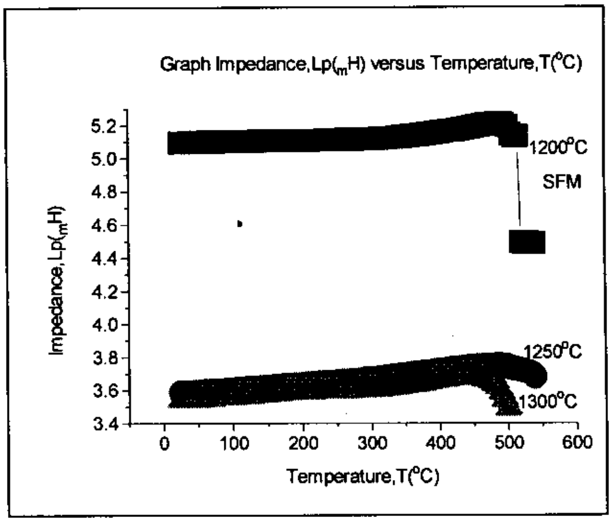


Fig.3: The Graph Shows the Curie Temperatures for SFM1200, SFM1250 and SFM1300

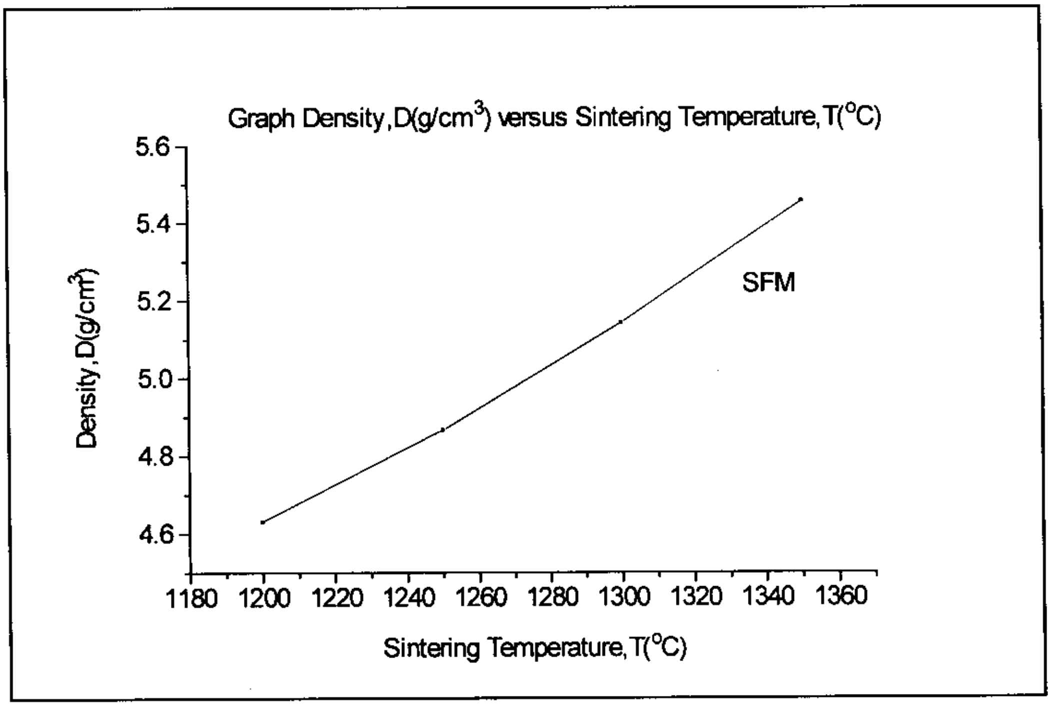


Fig. 4: The Relation of Increase Sintering Temperatures and Density Value of SFM1200, SFM1250 and SFM1300

using Archimedes principal. The Curie temperatures, T_c was obtained by plotting inductance against temperature.

Results and Discussion

The XRD patterns are shown in Fig.1. The FeO was oxidize at 400°C, 500°C and 600°C for 10 hours in air. Broad and clear diffraction lines begin to appear for powders oxidized at 400°C which correspond to progression of crystal growth of the entire particles. Sharp and clear diffraction lines pattern can be observed for powders oxidized at 600°C. The maximum duration to draft a sharp peak and pattern is 10 hours oxidation in air.

A close study on the effect of varying the sintering temperature on microstructure of strontium hexaferrite millscale derived has been done. Fig. 2 shows the SEM micrographs for sintering the strontium hexaferrite millscale derived at 1200°C, 1250°C, 1300°C and 1350°C. For sample SFM1200, fine-grained microstructure (about 0.3μm-0.7μm) was obtained due to the low sintering temperature employed in this work. The low density result, close to 90% of the theoretical value was also obtained for this sample (Fig. 4 and Table 1). Having closer look at sample, SFP1200 (Fig. 2 (e)) employing high purity Fe_2O_3 as the starting powder, a fine-grained microstructure was also obtained. By increasing temperature to 1300°C, large grains (5μm-7μm) were observed (Table 1). The increased sintering temperature, from 1200°C to 1300°C and 1350°C also resulted in higher density of the sample (Table 1 and Fig.4). The effect of the careful starting preparation methodology and the homogeneous powder mixtures is speculated as a main reason of the fine micro structure achieved. The control of sintering temperatures and duration of the process is thus important in hard ferrite fabrication.

Interesting however is the abnormal grain growth showed for $SrFe_{12}O_{19}$ sample (Fig.2 (d)). It is believed that the high sintering temperature (1300°C) and the existance of some impurities that acted as sintering aid, contained in the FeO (millscale), played an important role for the occurrence of the growth. It is speculated that these impurities and the high sintering temperature, provided fast diffusion path between solid grains, thus result in the abnormal grain growth. The absence of such growth during sintering at lower sintering temperature may support the above speculation.

Table 1: The Density Value, an Average Grain Size and Curie Temperature Result for the Samples

Sample	Density (g/cm^3)	Grain Size (μm)	T_c (°C)
SFM1200	4.6303	0.3-0.7	490
SFM1250	4.8662	0.9-1.2	465
SFM1300	5.1426	5-7	450
SFP1200	4.5632	0.3-0.8	495

The Curie temperatures of the sample SFM1200, SFM1250 and SFM1300 also resulted. The high sintering temperatures resulted a high density value (Fig. 4 and Table 1). A fine-grained, well-sintered samples found the nearly theoretical density. The effect of controlling the sintering temperature is the impediment for the micro structure development. This explains the high density and high quality of all samples. From the Fig. 4 shows that less than 1% of porosity appear and apparent as inter-granular pores. Here, we speculate the fine-grained microstructure and the low theoretical porosity would lead to a large BHmax value.

Conclusion

It could be concluded that the low-cost purification technique yielded highly purified wustite,FeO whose conversion to a very pure raw material Fe_2O_3 success to achieve approached as indicated by XRD peaks phase observation. A fine-grained and single-phase of $SrFe_{12}O_{19}$ millscale-derived have been achieved with a micro-particle size (0.3μm-0.7μm). The 1250°C sintering temperature for 10 hours in air is the maximum critical sintering temperature to avoid the occurrence of abnormal grain growth and the agglomeration of microstructure. Here, we conclude that the raw material composition of strontium hexaferrite forms a solid secondary phase during sintering which could prevent exaggerated grain growth. Also the starting powder morphology strongly governs this abnormal grain growth behaviour.

The effect of sintering temperature on orientation grain growth orientation in $SrFe_{12}O_{19}$ has been confirmed by the micro structure surface scan using SEM and was in good agreement with the XRD analysis based on the peak intensity of the (107) plane and also a high density was obtained. The high density value of $SrFe_{12}O_{19}$ was achieved with the increasing sintering temperature.

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