Effect of various parameters on the microstructure and magnetic properties of sintered Sr-hexaferrite

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ABSTRACT

Sr-hexaferrite powders with Fe/Sr molar ratio of 11 was synthesized via conventional ceramic route. Powders were milled for 5–25 h, then pressed to make green compacts by applying a pressure of 100–400 MPa. In other series, K2CO3 was used in starting materials. K2CO3/SrCO3 mixing molar ratio was 0.05 and 0.5. First series were calcined at 1250 °C, and the second series were calcined at 1000 °C for 2 h. The XRD characterization results revealed Sr-hexaferrite structure has been produced in both series; Crystallite size is constant then increases while lattice strain rises by variation of milling time from 5 h to 25 h. (BH)max and B increase by increasing milling time to 25 h and (BH)max reaches to optimized value of 24.75 kJ/m³. SEM was used for microstructure investigation; particle morphologies are hexagonal shape. Addition of slight amount of K2CO3 makes SrFe12O19 crystallites get less platelet shape and more regular shape. By increasing pressure morphology of the grain enhances in regularity. Magnetic properties were measured via permagraph; By increasing sintering temperature from 1150 °C to 1200 °C, density increases from 3.38 g/cm³ to 4.85 g/cm³. Maximum energy product varies from 9.54 kJ/m³ to 25.27 kJ/m³. Between the sintering temperatures of 1200–1250 °C maximum energy product stays at the highest values, But after 1250 °C drops to 3.82 kJ/m³. As density increases, remanence (Br) and maximum energy product [(BH)max] first increase at the pressure range of 100–200 MPa until it reaches to a optimize value of 15.20 kJ/m³ at 200 MPa.

Introduction

M-type strontium ferrites characterization and magnetic properties was first studied in 1950s by Philips [1]. This ferrite is magnetically hard with a high coercivity, although it has lower saturation magnetization in comparison with alloy magnets. Moreover it is much cheaper to produce. M type ferrites have high magnetic uniaxial anisotropy along the c-axis [2]. Nowadays, these ferrites have applications as recording materials, electromagnetic wave absorbing materials, dielectrics, rotors in permanent magnet motors, etc [3].

If a large coercivity is desired, the particle size should be in the order of the critical diameter dc for single domain behavior.

Hcj (intrinsic coercivity) and Hcb (extrinsic coercivity) increases by milling time as grain size decreases [4]. For Sr-ferrite, critical single domain particle size is dc ≈ 1 µm [5].

Sr-ferrites are synthesized by several technics such as: sol-gel method [6] hydrothermal method [7], coprecipitation method [8], self-propagating high temperature synthesis route [9], salt-melt method, conventional ceramic technique [10] and Low TemperatureCalcination Ceramic (LTCC) method. Salt-melt, conventional ceramic and LTCC methods are commonly used in industrial manufacturing. Also in salt-melt method agglomeration is less, thus pulverizing is not needed after calcination [11]. Sr-ferrites were synthesized by salt-melt method using Na2CO3 and Na2SO4 as a flux and Na+/Sr2- molar ratio were selected 1.3 and 0.2, respectively [5,12]. Attempts to reduce calcination temperature with much less amount of flux, LTCC technology was performed by using BaCu(B2O5) flux in range of 1–4 wt% [13], and in another study, K2CO3 flux was used with K+ /Sr2- molar ratio, in range of 0.01–0.02 [11]; Moreover, LTTC method reduces the calcination temperature, besides the washing stage after calcination is not required unlike salt melt method, since low amount of flux is added.

In these magnets, effect of various parameters on magnetic properties have been tried. Some of these parameters increase both coercivity and magnetic remanence (Br), some of parameters increase just coercivity and some others increase just magnetic remanence. For a large (BH)max we need to attain both high Br and Hc parameters [14]. By this mean, production of a dense microstructure and less grain growth is needed. For this purpose several ways like doping with cations like La-Co [15], using sintering aids like Bi2O3 [16], Ca-Si [17,18] and Al2O3 has been investigated [19,20]. Also, Effect of various
parameters like: sintering and calcination temperature and the milling time have been investigated on magnetic properties of Sr-ferrites [21–23]. Influence of pressing compression on density is investigated for other composites [24,25]. Pulse magnetic field pressing was simulated in previous studies [26,27]. Particles size get larger in sintering so milling before sintering is a way to constrain tremendous growth of some grains, by reducing grain size to under \(d_0\) (1 \(\mu\)m), before sintering stage. Effect of milling time on particle size and magnetic properties of ferrite has been investigated [28,29].

In a research Sr-hexaferrite magnets with high performance were achieved by addition of 0.5%wt SiO\(_2\) and 0.5%wt CaCO\(_3\) as sinter-aids [18]. Heretofore effect of milling time and K\(_2\)CO\(_3\) addition on lattice strain of Sr-ferrite has not been investigated. As well comparing different amount of K\(_2\)CO\(_3\) which alters synthesize method from LTTC route to salt-melt has not been examined. Increasing pressure would be a low cost method for increasing magnetic properties. On the other

| Parameters | Properties |
|------------|------------|
| Sample | Applied pressure (MPa) | Milling time (h) | K\(_2\)CO\(_3\)/SrCO\(_3\) Molar ratio | Sintering temperature (°C) | Density (g/cm\(^3\)) | \(B_r\) (mT) | \(H_{cb}\) (kA/m) | \(H_{cj}\) (kA/m) | \((BH)_{max}\) (kJ/m\(^3\)) | Sintered magnet diameter (mm) | Radial shrinkage (%) |
| P1 | 100 | 5 | 0 | 1250 | 4.33 | 298 | 167.83 | 190.51 | 15.20 | 14.6 | 14.12 |
| M1K0P2 | 200 | 5 | 0 | 1250 | 4.44 | 329 | 178.17 | 191.07 | 19.26 | 14.84 | 12.71 |
| P3 | 300 | 5 | 0 | 1250 | 4.53 | 355 | 185.42 | 195.60 | 22.68 | 15 | 11.76 |
| P4 | 400 | 5 | 0 | 1250 | 4.64 | 365 | 191.78 | 207.86 | 24.11 | 15.3 | 10 |
| K1 | 200 | 5 | 0.05 | 1250 | 4.56 | 289.00 | 84.27 | 107.43 | 7.00 | 13.5 | 20.59 |
| K2 | 200 | 5 | 0.5 | 1250 | 4.6 | 348.00 | 135.28 | 153.58 | 16.00 | 13.41 | 21.12 |
| M2 | 200 | 10 | 0 | 1250 | 4.67 | 329 | 178.17 | 191.07 | 19.26 | 14.36 | 15.53 |
| M3 | 200 | 15 | 0 | 1250 | 4.68 | 355 | 185.42 | 195.60 | 22.68 | 14.2 | 16.47 |
| M4 | 200 | 20 | 0 | 1250 | 4.81 | 365 | 191.78 | 207.86 | 24.11 | 14.2 | 16.47 |
| M5 | 200 | 25 | 0 | 1250 | 4.86 | 370 | 183.19 | 196.72 | 24.75 | 14.6 | 14.12 |
| T1 | 200 | 20 | 0 | 1150 | 3.38 | 225.00 | 165.36 | 306.13 | 9.55 | 15.5 | 8.82 |
| T2 | 200 | 20 | 0 | 1200 | 4.85 | 367.00 | 186.77 | 196.80 | 24.27 | 14.5 | 14.71 |
| T3 | 200 | 20 | 0 | 1250 | 4.75 | 368.00 | 187.25 | 197.03 | 24.75 | 14.6 | 14.12 |
| T4 | 200 | 20 | 0 | 1300 | 4.84 | 320.00 | 43.77 | 50.93 | 3.82 | 14.4 | 15.41 |

Fig. 1. XRD patterns of Sr-hexaferrite as-synthesized powders for samples (a) M1K0P2, (b) M2, (c) M3, (d) K1 and (e) K2 (* symbolized peaks are for α Fe\(_2\)O\(_3\) and unsigned peaks are for Sr-hexaferrite.) with Williamson-Hall plots (for 14 intense peaks) and Williamson-Hall equations: \(y = −0.00x + 0.01\), \(y = −0.003x + 0.0052\), \(y = 0.0014x + 0.003\), \(y = −2×10^{-5}x + 0.0046\) and \(y = −2×10^{-5}x + 0.004\), for each of the specimens, respectively.
hand, the effect of sintering temperature at presence of (CaO + SiO2) sinter-aid on magnetic properties is going to be discussed in optimized pressure and milling time.

In this study, effect of these parameters by the addition of the 0.5% wt SiO2 and 0.28%wt CaO as sinter-aids on sintered SrFe11O19 magnets are investigated to reach maximum magnetic properties.

### Table 2
Structure parameters, theoretical density ($\rho_x$), measured density ($\rho_m$), porosity (P) and geometry parameters for the as synthesized samples M1K0P2, M3, M5, K1 and K2 before sintering.

| Sample | Milling Time | $K_2CO_3/SrCO_3$ molar ratio | a (nm) | c (Å) | c/a ratio | Cell Volume ($nm^3$) | $\rho_x$ (g/cm$^3$) | $\rho_m$ (g/cm$^3$) | P (%) | Crystallite Size(nm) | Strain |
|--------|--------------|------------------------------|--------|------|-----------|----------------------|----------------------|----------------------|-------|---------------------|--------|
| M1K0P2 | 5            | 0                            | 5.882  | 23.050 | 3.919     | 690.591              | 5.106                | 4.44                 | 13.04 | 27.18               | $-5.00 \times 10^{-4}$ |
| M3     | 15           | 0                            | 5.880  | 23.039 | 3.918     | 689.772              | 5.112                | 4.68                 | 8.45  | 26.65               | $-3.00 \times 10^{-4}$ |
| M5     | 25           | 0                            | 5.884  | 23.051 | 3.918     | 691.098              | 5.102                | 4.86                 | 4.75  | 46.20               | $1.40 \times 10^{-3}$  |
| K1     | 5            | 0.05                         | 5.890  | 23.072 | 3.917     | 693.217              | 5.087                | 4.56                 | 10.35 | 30.13               | $-2.00 \times 10^{-5}$ |
| K2     | 5            | 0.5                          | 5.878  | 23.015 | 3.915     | 688.591              | 5.121                | 4.43                 | 13.49 | 32.23               | $-2.00 \times 10^{-5}$ |

**Fig. 2.** SEM micrographs of samples (a) M1K0P2, (b) M4, (c) K1 and (d) K2, (by ×2000). And SEM images for particle morphologies of samples M1K0P2 and K2 (by ×20000).

### Experimental

The starting materials; SiO2, CaO, SrCO3 (99% purity, Sigma Aldrich) and Fe2O3 from steelmaking by-product (93% purity) were used. For synthesizing Sr-hexaferrite starting materials were mixed with a Fe2O3: SrCO3 molar ratio of 5.5 in a planetary ball mill. The weight
ratio of the ball to powder was 10:1 and mixture was wet milled in distilled water for 2 h. Powder mixture was dried and calcined at 1250°C for 2 h, in a muffle furnace. The calcined powders were pulverized in a marble mortar and sifted to under 100 µm particle size. Sr-ferrite powders with 0.28% wt CaO, 0.5% wt SiO$_2$ additives were wet milled in a planetary ball mill, at a speed rate of 200 rpm, for 5–25 h by 5 h increment, for samples M1K0P2-M5, respectively. The finely milled slurry for 5 h was pressed into disk shape compacts with diameters of 17 mm and thickness of 4–7 mm by wet molding in a 800 kA/m magnetic field which was parallel to the pressing direction (sample P1) under 100 MPa. After turning off magnetic field, further pressing to 200, 300 and 400 MPa was performed for samples M1K0P2, P3 and P4, correspondingly. Green compacts were sintered in a muffle furnace at 1250°C for 15 min. To investigate the effect of sintering temperature, some of compacts were sintered at 1150°C, 1200°C, 1250°C and 1300°C named as samples T1, T2, T3 and T4, respectively.

Meanwhile effect of K$_2$CO$_3$ addition was investigated by mixing formula SrCO$_3$, 5.5Fe$_2$O$_3$, nK$_2$CO$_3$ in which n = 0.05 and 0.5 are referred to samples K1 and K2, respectively. Process of the making these magnets was as same as the above mentioned ceramic route ones, unless Powder mixture was calcined in a muffle furnace at 1000°C for 1 h. The calcined powder was mixed with 10% nitric acid in boiling water and then it was rinsed with distilled water this was repeated until the color of the solution didn’t change. Washing process for sample K2 was not necessary. The rest of process was done as same as first series of samples. The radial shrinkage, density and magnetic properties were measured and listed in Table 1. X-ray diffraction patterns of the magnetic powders were carried out by X-ray diffractometer, with Cu K$_\alpha$ radiation source ($\lambda = 1.5402\ \text{Å}$), Siemens D 5000. Microstructure of the sintered compacts was investigated using a field emission scanning electron microscope (FEG SEM), MIRA3-TESCAN. Furthermore, magnetic properties of magnets were measured utilizing a permagraph C500, Magnet-Physic Steingroover GmbH, under 800 kA/m applied field at room temperature.

Results and discussion

Effect of milling time and synthesis method

Fig. 1 exhibits existence of trace amount of Fe$_2$O$_3$ phase, which is attributed to saturated Fe$^{3+}$ at different crystallographic positions. It can be concluded that the iron content (x) plays an important role in the formation of the single-phase hexagonal ferrite, for which iron content is lower than stoichiometric ratio, whereas stoichiometric ratio even

![Fig. 3](image-url)
less than 5.5 is needed [30].

By increasing milling time from 5 in Fig. 1(a) to 15 h in Fig. 1(b), the intensity of peak (0 0 8) in respect to other peaks reduces, then increases again at 25 h in Fig. 1(c). Decreasing a (X00) peak intensity in respect to other peaks means c/a aspect ratio of crystal parameters lessens meaning grains get platelet shape or with less width [31].

In Fig. 1(d) and (e), XRD patterns show peak intensity increases by raising K2CO3/SrCO3 molar ratio. This is due to completion of formation of Sr-ferrite phase by either increasing K2CO3/SrCO3 molar ratio or calcination temperature [5,32]. It is clear from Fig. 1(d) and (e) under the influence of c axis, relative intensity of (0 0 8) peak in respect to other peaks is elevated by increasing K2CO3/SrCO3 molar ratio, owning to the grain growth along c axis [33].

Also it shows that by increasing the milling time, the width of Sr-ferrite diffraction peaks broadens and their intensity gradually decreases. Williamson-Hall equation was used to measure lattice strain and grain size of the calcined and milled powders [34]. Williamson-Hall plots in Fig. 1(a), (b) and (c) show by increasing milling time from 5 h to 15 h lattice strain obtained from Williamson-Hall plot increases from −5.00 × 10−4 to −3.00 × 10−4; in which lattice strain is proportion to the slope of the line in Williamson-Hall equation, then by increasing milling time to 25 h it increases to a compression strain of 1.40 × 10−3.

By increasing the milling time, grain size decreases slightly and lattice strain increases. By further increasing of the milling time, strain is enhanced and also grain size increases. Grain size increasing is as a result of strong inter-particle aggregation promoted by intensive milling. A similar aggregation phenomenon has been observed in other research and different materials after prolonged milling time [35,36]. Milling method causes defect, lattice distortion and reduction of grain size which provides an additional energy into milling powder, so strain is increased [37]. XRD in Fig. 1(d) and (e) show that in ceramic route, there is a little tensile strain, with increasing milling time, strain transfers from tensile to compression.

It is clearly understood from Table 2, crystals have lower strain than solid state reaction, this can be owing to Sr-ferrite grains formation in a liquid phase media of K2CO3, in salt-melt method [38]. It is obvious from Fig. 1(a), (d) and (e) crystallite size increases by K2CO3/SrCO3 molar ratio, from 27.18nm in solid state route (M1K0P2) to 30.13 nm in LTCC route (K1) and then to 32.23 nm in salt-melt (K2) method.

Lattice parameters ‘a’ and ‘c’ are calculated from (1 1 4), (0 1 7)
peaks. X-ray diffraction data is used to estimate the X-ray density ($\rho_x$) of magnetic powders and 'M' refers to molar mass. The bulk density ($\rho_m$) and volume ($V$) is measured from Archimedes Principle and M refers to molar mass. The porosity ($P$) of the magnets is also calculated. The following equations are used for calculating the parameters in Table 2.

$$V = 0.8666a^2c$$  \hspace{1cm} (1)

$$\rho_x = 3.321 \times 10^{22} \frac{M}{V}$$  \hspace{1cm} (2)

$$P = \left(\frac{\rho_x - \rho_m}{\rho_x}\right) \times 100$$  \hspace{1cm} (3)

Verstegen and Stevels concluded the c/a parameter ratio determines structure type, as for M-type structure, this ratio must be lower than 3.98. According to Table 2, M type structure is formed in synthesizing by either of the techniques [39].

As it is seen from Fig. 2(c) and (d) in salt-melt method particles are smaller, have homogenized and spherical shape in compared with ceramic route in Fig. 2(a) and (b) which has irregular shape and agglomerated particles [13]. It is observed in Fig. 2(a) and (b), by increasing milling time from 5 h to 20 h, grains size decrease from more than 1 µm to about 1 µm. And Fig. 2(b) and (c) shows by increasing $K_2CO_3/SrCO_3$ molar ratio from 0.05 to 0.5 grain size increases, however no abnormal grain growth happens and particle size distribution remains narrow.

According to a study performed by F. Kools et al. [40], the intrinsic coercivity could be expressed by the following equation:

$$H_{cj} = aH_a - \frac{N(J_s + J_c)}{\mu_0}$$  \hspace{1cm} (4)

where 'a' is the grain size factor, 'H_a' is the magnetocrystalline anisotropy field, 'N' is the grain demagnetization factor and 'J_s' is the saturation polarization of the magnet. The factor a increases with the decreasing of grain size; the factor N increases when the grain shape becomes more platelet. Moreover, the intrinsic coercivity of the ferrite increases with both decreasing the grain size and lowering platelet shape of hexagonal grains, simultaneously.

Remanence is calculated from the equation below in which the parameter '$\rho_m\$' is density, 'c' is the anisotropy factor and '$J_s$' is the saturation polarization.

$$B_r = J_s \rho_m$$  \hspace{1cm} (5)

If both quantities ($\rho_m$ and $\alpha$) for a sintered microstructure approach the highest value, maximum remanence will be reached. Therefore, $B_r$ increases with the enhancement of density [34].

By increasing milling time from 5 h to 25 h, density increases; it could be because of reduction of grain size which employs high surface energy to motivate force for sinterability (Fig. 3).

Firstly, reducing particle size raises coercivity from 167.83 kA/m at 5 h to 191.78 kA/m at 20 h of milling time. Secondly, According to Williamson-Hall equation, increasing of the milling time raises lattice strain (Fig. 1(a), (b) and (c)). According to previous works, increasing lattice strain could also lead to coercivity enhancement [41]. On the contrary, as it is seen in Fig. 3(c), radial shrinkage decreases after 15 h milling, while density increases. So axial shrinkage raise, i.e. grain growth occurs perpendicularly to c axis, parameter a increase, parameter N increases as well. Consequently, after a while parameter N outweighs parameter a which causes $H_{cj}$ decrease after 20 h (Fig. 3(d)).

$$(B\cdot H)_{max}$$ is the maximum surface area under demagnetization curve and $H_{cj}/H_{cj}$ indicates the rectangularity of the demagnetizing curves for the magnets so it has the same trend with $(B\cdot H)_{max}$ [42]. It is seen from Fig. 3(e), $(B\cdot H)_{max}$ improves by increasing milling time from 15.20 kJ/m$^3$ at 5 h to 24.75 kJ/m$^3$ at 25 h.

Comparing the radial shrinkage and density values in Fig. 4(c), demonstrates that with addition of $K_2CO_3$, radial shrinkage falls to a lower amount in compared with M1K0P2 magnet whereas density increases, ensuring axial shrinkage is lower in salt melt and LTCC technique so particles grow preferentially along c axis (getting less platelet shape), in sintering stage. On the other hand, in Fig. 1(a), (b) and (c) it is observed that in as synthesized Sr-ferrite powders for sample K1 grains are platelet shape in comparison to M1K0P2 and K2 magnets, which is a factor for rise in N and reduction in $H_{cj}$ from 167.83 kA/m for M1K0P2 magnet to 84.27 kA/m for K1 magnet (according to Eq. (4)). It increases up to 135.28 kA/m for sample K2 which could be as a
result of getting less irregular and more spherical, as Fig. 2(b) and (c) demonstrate, by addition of more $K_2CO_3$. It is also explained by the increase of particle size which in turn provides transition from the super-paramagnetic behavior to the normal state of the single domain nanoparticles (as it is seen in Fig. 2(d), particle size is under $d_p = 1 \mu m$ for salt-melt method).

As it is seen from Fig. 2(b) and (c) samples K1 and K2 with salt-melt method are synthesized in low temperature so they have finer and near spherical shape powders which assist to more density and this increases by increasing flux to powder ratio. So, $B_r$ does not change a lot from 298 in M1K0P2 magnet to 289 mT in K1 magnet and then increases to 348 mT in K2 magnet.

It is seen in Fig. 4(e), $(BH)_{max}$ decreases from 15.20 kJ/m$^3$ in solid state reaction synthesizing method to 7 kJ/m$^3$ in LTCC route, then increases to 16 kJ/m$^3$ in salt-melt route.

**Effect of sintering temperature**

Fig. 6(d) shows variation of intrinsic and extrinsic coercivity by versus sintering temperature. For Sr-ferrite magnets sintered at 1150 °C, Mean particle size is 0.46 µm with 0.16 µm standard deviation (Fig. 5(a)), until mean particle size increases by increasing sintering temperature, contributing to increase in the value of $H_c$ from 165.36 kA/m at 1150 °C to 187.25 kA/m at 1250 °C which grains grow to about single domain particle size (Fig. 5(b)), then after reaching to mean particle size more than 1 µm at sintering temperature of 1300 °C (Fig. 5(c)), coercivity decrease to 43.77 kA/m [5]. Fig. 5(d) shows oriented platelet like morphologies in the surface of magnet, parallel to the pressing direction [12].

(CaO + SiO2) sinter-aid and excess amount of SrO exist together, at grain boundary. Thus it is seen in Fig. 6(c) Density increase sharply, for the temperature ranging from 1150 °C to 1200 °C [18]. Then it remains about constant up to 1300 °C. In Fig. 6(d), $B_r$ parameter increases by increasing temperature as a result of density enhancement. Next, it starts decreasing after 1200 °C, this might be owing to the formation of multi-domain and the easy movement of the domain walls, resulting in domains misalignment that decreases anisotropy factor in equation (5) and consequently $B_r$ decreases [45].

It is clearly understood from Fig. 6(e), $(BH)_{max}$ mounts by increasing sintering temperature from 9.55 kJ/m$^3$ at 1150 °C to 24.27 kJ/m$^3$ at 1200 °C. It reaches the maximum value of 24.75 kJ/m$^3$ at 1250 °C. Then starts decreasing to 3.82 kJ/m$^3$ at 1300 °C.

![Fig. 6. Demagnetization curves collected for sintered Sr-ferrites samples T1, T2, T3 and T4 with (a) Hcb and (b) Hcj versus magnetic remanence. Effect of sintering temperature on (c) density, radial shrinkage, (d) $B_r$, $(BH)_{max}$ and (e) coercivity.](image)
**Effect of pressing pressure**

SEM micrographs in Fig. 7 show grain grows perpendicular to c axis. And particle size increases by compression from 0.44 µm in 100 MPa to 1.53 µm in 400 MPa and the grain becomes more platelet shape. with standard deviation decreasing from 0.65 to 0.78, so particle size distribution gets broader and shifts to the left.

Fig. 8(a) demonstrate by applying more pressure density improves while radial shrinkage decreases, in order to make balance, apparently axial shrinkage must increase to counteract radial shrinkage influence on density which makes grain get platelet shape. As it is seen in Fig. 7, by increasing pressure, particles get more regular platelet like hexagonal shape in regard to samples pressed at 100 MPa in Fig. 7(a) which is in agreement with above mentioned particle shape variation using Fig. 8(a). Also particles size gets even larger than 1 µm in Fig. 7(b) SEM micrographs. These occurring make factor N increase and factor a decrease, Hence Hcj decreases.

According to Fig. 8(c) and (d), by increasing pressure, density of the specimens enhances that leads to increasing of the B, from 284 mT at 100 MPa to 298 mT at 200 MPa, However, it does not change a lot when the pressure is between 200 and 300 MPa, but again increases to 305 mT at 400 MPa. Since (BH)max, maximum surface area surrounded under Hc versus Br plot, is directly proportional with both B, and Hc, So here with these parameters not having akin inclination, (BH)max has Two optimaums, one at 200 MPa and the other one at 400 MPa.

Fig. 8(e) shows, as milling time increases, maximum energy product [(BH)max] first increase from 13.68 kJ/m³ at the pressure of 100 MPa to optimum value of 15.20 kJ/m³ at the pressure of 200 MPa then decreases to 14.40 kJ/m³ at 300 MPa then rises again to 14.96 kJ/m³ at 400 MPa.

**Conclusions**

Sr-hexaferrite was synthesized via solid-state reaction route which contains little amount of Fe₂O₃. According to the XRD results, Sr-hexaferrite has been produced; the crystallite size range was 27–46 nm; it is about constant from 5 h to 15 h, then increases, while lattice strain rises by milling time. Magnetic properties were studied via permagraph. Furthermore, Hc and Br enhance with rising the milling time. However, coercivity decreases after 20 h milling, i.e. there is an optimum time of milling for achieving the maximum Hc. The range of Hcj rises from 167.83 to 191.78 kA/m. besides, Hcj increases from 167.83 to 191.78 kA/m, during the milling time varying in the range of 5–20 h.

Addition of K₂CO₃ makes SrFe₁₂O₁₉ crystallites get smaller in size. (BH)max decreases from 15.20 kJ/m³ in ceramic route to 7.00 kJ/m³ in LTCC route and increases to 16 kJ/m³ in salt-melt route. Likewise, remanence, extrinsic coercivity and intrinsic coercivity have the same trend with (BH)max by increasing addition of K₂CO₃ flux amount. As each of these three magnetic parameters, increase by increasing either of calcination temperature or K/Sr molar ratio.

By increasing sintering temperature from 1150°C to 1200°C, density increases from 3.38 g/cm³ to 4.85 g/cm³, then remains about the same value. (BH)max varies from 9.54 kJ/m³ to 25.27 kJ/m³. Between 1200 and 1250°C maximum energy product stays at the highest value, then after 1250°C drops to 3.82 kJ/m³ at 1300°C. Similarly, Br reaches to optimum value ranges of 367–368 mT at sintering temperature range.
of 1200–1250 °C and falls by increasing temperature to 1300 °C, as multi-domain particles form. Nevertheless, coercivity decreases by sintering temperature from 1150 to 1300 °C.

By increasing pressure, morphology of the grain enhances in platelet shape and larger in size. \( H_c \) and \( B_r \) have the inverse and direct proportion with the compression quantity (100–400 MPa), respectively; except pressure range of 200–300 MPa in which the \( B_r \) does not increase with enhancement of the pressure. Since \( (BH)_{\text{max}} \) has directly proportional with \( B_r \) and \( H_c \), \( (BH)_{\text{max}} \) has Two optimums on at 200 MPa and the other one at 400 MPa.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rinp.2019.01.038.

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Fig. 8. Demagnetization curves collected for sintered Sr- ferrites samples P1, M1K0P2, P3 and P4 with (a) \( H_{cb} \) and (b) \( H_{cj} \) versus magnetic remanent. Effect of pressing pressure on (c) density, radial shrinkage, (d) \( B_r \), (BH)\text{max} and (e) coercivity.

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S. Garehbaghi, A. Kianvash

Results in Physics 12 (2019) 1559–1568

1567
