

Low Temperature Sintering Characteristics of Hot Press Sintered $\text{SrFe}_{12}\text{O}_{19}$ Ferrites for Use in Microwave LTCC Circulators



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Abstract: The $\text{SrFe}_{12}\text{O}_{19}$ ferrites with different amounts of Bi_2O_3 additive were prepared by a hot press sintering process at a low fired temperature of 870 °C in order to be compatible to the LTCC (low temperature co-fired ceramics) technology, and their low temperature sintering characteristics were investigated, including the crystal phase composition, sintering density, porosity, and magnetic properties. Results show that the addition of Bi_2O_3 promotes the formation of $\text{SrFe}_{12}\text{O}_{19}$ phase structure and increases the sintering compactness and magnetic properties of the ferrites fabricated at 870 °C. The ferrites with Bi_2O_3 content from 2 wt% to 4 wt% exhibit a compact microstructure with sintering density higher than $4.65 \text{ g}\cdot\text{cm}^{-3}$ and porosity lower than 10%, which contributes to the enhanced saturation magnetization M_s and intrinsic coercivity H_{ci} above $252.4 \text{ kA}\cdot\text{m}^{-1}$ and $312.9 \text{ kA}\cdot\text{m}^{-1}$, respectively. Moreover, the potentiality of the $\text{SrFe}_{12}\text{O}_{19}$ ferrites for use in microwave LTCC circulators was also discussed based on their low temperature sintering characteristics.

Key words: $\text{SrFe}_{12}\text{O}_{19}$ ferrites; LTCC; hot press sintering; circulators

Low temperature sintering of gyro-magnetic ferrites is a crucial problem for microwave LTCC (low temperature co-fired ceramics) ferrite devices^[1-6]. Because of high sintering temperature, the traditional and commercial gyro-magnetic ferrite materials are unable to co-fire with the inner electrode metal Ag in LTCC systems. M-type hexagonal barium ferrites ($\text{BaFe}_{12}\text{O}_{19}$) and strontium ferrites ($\text{SrFe}_{12}\text{O}_{19}$) can be used in microwave and millimeter wave ferrite devices due to their relatively high saturation magnetization M_s , moderate intrinsic coercivity H_{ci} , large crystalline anisotropy field H_a , and appropriate resistivity ρ , which have been considered as the promising microwave LTCC ferrite materials^[7-9]. Different additives were used as sintering aids to reduce the sintering temperature of the ferrites, typically including Bi_2O_3 , BaCu (B_2O_5) and $\text{Bi}_2\text{O}_3\cdot\text{B}_2\text{O}_3\cdot\text{SiO}_2\cdot\text{ZnO}$ (BBSZ), and their low

temperature sintering characteristics were improved^[10-12].

In the present paper, the $\text{SrFe}_{12}\text{O}_{19}$ ferrites with different amounts of Bi_2O_3 additive were prepared by a hot press sintering process at a low fired temperature in order to be compatible to the LTCC technology. The crystal structure and magnetic properties of the ferrites were mainly investigated, and their potential applications in microwave LTCC circulators were also discussed.

1 Experiment

High purity ($\geq 99.99 \text{ wt\%}$) SrCO_3 and Fe_2O_3 were weighed in composition of $\text{SrFe}_{12}\text{O}_{19}$ for preparing stoichiometric ferrites. Raw materials were mixed and ball-milled at 300 r/min for 6 h, then dried and calcinated at 1250 °C for 2 h. Powders were further ball-milled with different amounts of Bi_2O_3 additive from 0 to 5 wt% at 400

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r/min for 4 h, and then pressed into disks using a graphite mold and sintered at 870 °C under 60 MPa for 10 min. Particle size of powders was tested by laser particle analyser (JL-1178). Sintering density was measured by precision density balance (FA2004J) with resolution of 0.1 mg based on the Archimedes method. Crystal structure was detected by X-ray diffraction (XRD, DX-2700) with Cu K α radiation. Direct-current resistivity was measured by precision power supply (Agilent, B2912A) from 0 to 200 V with resolution of 10 fA/100 nV. Magnetic hysteresis loops were tested by vibrating sample magnetometer (VSM, Versalab) to obtain the magnetic properties.

2 Results and Discussion

XRD patterns of the strontium ferrites with different Bi₂O₃ contents are shown in Fig.1. The SrFe₁₂O₁₉ phase with M-type hexagonal structure is formed in the ferrites with different Bi₂O₃ contents. For the ferrites without Bi₂O₃ additive, the SrFe₁₂O₁₉ phase coexists with the non-magnetic α -Fe₂O₃ phase and SrO₂ phase, where the diffraction peaks of α -Fe₂O₃ phase like (110), (020), (130) peaks and a weak (002) peak of SrO₂ phase are observed. This is in agreement with the formation mechanism of SrFe₁₂O₁₉ ferrites. The formation of SrFe₁₂O₁₉ ferrites prepared by a solid phase method is mainly based on the reaction as $\text{SrCO}_3 + 6\text{Fe}_2\text{O}_3 = \text{SrFe}_{12}\text{O}_{19} + \text{CO}_2\uparrow$, and the sintering temperature usually reaches above 1200 °C. Generally, Fe₂O₃ can not fully participate in the reaction to produce SrFe₁₂O₁₉ at low sintering temperatures. When the content of Bi₂O₃ increases to 1 wt%, the crystal phase composition of the ferrites is still unchanged. It is worthwhile to note that the SrFe₁₂O₁₉ ferrites with single phase structure are successfully obtained when the Bi₂O₃ content increases to 3 wt%. The strength of (006) peak and (008) peak is obviously enhanced, suggesting an increased *c* axis preferred orientation in the ferrites. Redundant addition of Bi₂O₃ additive is observed in the ferrites when the Bi₂O₃ content further increases to 5 wt%, where a small amount of Bi₂O₃ phase with weak (112), (121), and (012) peaks is detected. Moreover, the inter-planer spacing d_{hkl} of most crystal faces is found to get a significant decrease, which is strongly correlated with the lattice constants *a* and *c*.

Lattice constants of the strontium ferrites are calculated from the values of d_{hkl} corresponding to (107) peak and (114) peak by the following equations:

$$d_{hkl} = \left(\frac{4}{3} \frac{h^2 + hk + k^2}{a^2} + \frac{l^2}{c^2} \right)^{-1/2} \quad (1)$$

and the porosity *P* of the ferrites is obtained from

$$P = 1 - \frac{d_s}{d_x} \quad (2)$$

$$d_x = (1-x)d_M + xd_A \quad (3)$$

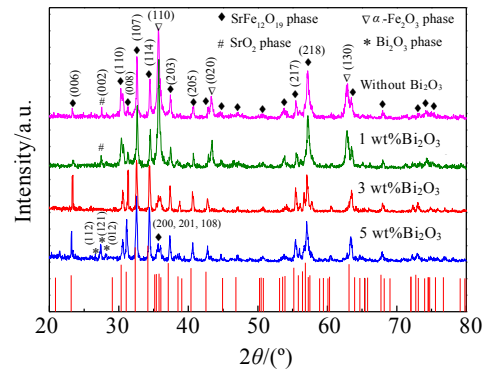


Fig.1 XRD patterns of the SrFe₁₂O₁₉ ferrites with different Bi₂O₃ contents

$$d_M = \frac{2M}{NV} \quad (4)$$

$$V = 0.8666a^2c \quad (5)$$

where *h*, *k*, and *l* are the Miller indices, d_x is the theoretical density of the ferrites, d_s is the sintering density of the ferrites, *x* is the amount of the additive, d_M is the X-ray density of SrFe₁₂O₁₉, d_A is the theoretical density of the additive, *M* is the molar mass of SrFe₁₂O₁₉, *V* is the lattice volume, and *N* is the Avogadro's number.

Evident effect of the Bi₂O₃ content on the lattice constants of the strontium ferrites is observed, as shown in Fig.2. The large lattice constant *a* from 0.5898 nm to 0.5922 nm is obtained with Bi₂O₃ content from 1 wt% to 4 wt%, which clearly exceeds the typical value of 0.588 nm. Correspondingly, the lattice constant *c* decreases to a range between 2.196 and 2.226 nm from a typical value of 2.307 nm. This indicates that the (001) face of the hexagonal structure is chiefly perpendicular to the pressure direction, leading to the lattice deformation.

Fig.3 gives the sintering density and porosity of the strontium ferrites with different contents of Bi₂O₃ additive. The addition of Bi₂O₃ is found to be conducive to increase the sintering density and to decrease the porosity of the ferrites, which is suggested to be strongly associated with the formation of Bi₂O₃ liquid phase during the sintering process. The ferrites can provide a relatively compact microstructure with sintering density higher than 4.65 g·cm⁻³ and porosity lower than 10% when the Bi₂O₃ content varies from 2 wt% to 4 wt%. Low porosity can reduce the demagnetization field inside the ferrites and decrease the electromagnetic loss in high frequency applications.

Moreover, the enhanced saturation magnetization M_s and intrinsic coercivity H_{ci} are obtained for the ferrites due to the promoted low temperature sintering with appropriate amount of Bi₂O₃ additive, as shown in Table 1. To the contrary, the redundant non-magnetic Bi₂O₃ phase results in depressed magnetic properties, and there is a great

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