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Microstructure and magnetic properties of NiZn ferrite prepared by ultrafast pulse electric current sintering without pressure

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Abstract: Polycrystalline nickel-zinc ferrite ceramics were prepared within tens of seconds via ultrafast pulse electric current sintering without pressure. The phase composition and microstructure of the samples were characterized by X-ray diffraction and scanning electron microscopy, respectively. The static magnetic properties and Curie temperature of the samples were investigated by vibrating sample magnetometry. The main phase of the sintered sample was the same as that of the raw powder, and the maximum relative density of the sample reached 95.72%. The densification and grain growth of the sample were accelerated through the synergistic effect of the energy concentration of pulse current heating and Joule heating that was generated by the sample. The grains at the edge of the sample were larger than those at the center, which was attributed to the uneven temperature distribution in a sintering sample. The slight differences in specific saturation magnetization and Curie temperature of the samples were attributed to the slight difference in sample phase composition that was caused by partial nickel-zinc ferrite decomposition and Zn volatilization during sintering. The inferred critical grain size between the dual- and mono-domain was ~450 nm. The ultrafast pulse electric current sintering technique has the advantages of a heating energy concentration, high heating rate and energy saving. The technique provides an effective approach to prepare compact and ultrafine grain ceramics rapidly.

Key words: nickel-zinc ferrite; ultrafast pulse electric current sintering; Joule heating; rapid densification

1 Introduction

Ferrite is an oxide that contains iron and one or more other metals. The nickel-zinc (NiZn) ferrite with spinel structure is one of the typical representatives of soft magnetic materials. Because NiZn ferrite has the advantages of a simple preparation, high resistivity, low eddy current loss and good chemical properties, it is widely used in the telecommunications and high frequency power industries, such as inductors, transformers, isolators, magnetic amplifiers and transfer switches [1-5]. The magnetic properties of the ferrite depend on its chemical composition and microstructure [6,7]. In the spinel ferrite unit cell, tetrahedrons and octahedrons are formed by regular cubic close packing of the oxygen ions. NiZn ferrite is a mixed spinel, in which Zn^{2+} ions occupy the tetrahedral (A) sites, Ni^{2+} ions occupy the octahedral (B) sites, and Fe^{3+} ions occupy the A- and B-sites [8]. The magnetic moment of the A-sublattice is antiparallel to that of the B-sublattice. The magnetic moment of the NiZn ferrite is determined from the difference of the magnetic moments between the A- and B-sublattices [9]. The Curie temperature (T_c) of the NiZn ferrite depends on the superexchange interaction between the A- and B-sublattices. In addition, the NiZn ferrite microstructure has an important influence on the coercivity (H_c), ferromagnetic resonance linewidth, and permeability of the NiZn ferrite [5].

NiZn ferrite is commonly used in devices as ceramics that are processed into a certain shape. Sintering is a key step in ceramic preparation, which influences the ceramic microstructure. The conventional ceramic technique with a high temperature and long holding time increases the energy consumption significantly [10-12]. Furthermore, it leads to the volatilization of some components, which results in a composition instability and ferrite performance uncertainty because of the long holding time at a high temperature. To shorten the sintering time and/or to reduce the sintering temperature, extensive attention has been focused on the ceramic sintering technique. To date, many techniques, such as spark plasma sintering [13-17], electric current/field-assisted sintering [18-23], hot pressing sintering [24-26] and microwave sintering [27-32], are used for this purpose.

In this work, NiZn ferrite ceramics were prepared through ultrafast pulse electric current sintering (UPECS) without pressure. The raw material was $\text{Ni}_{0.75}\text{Zn}_{0.25}\text{Fe}_2\text{O}_4$ nanopowder without any additives. During the UPECS process, the sample was kept above 600°C for no more than 90 s.

The microstructure and magnetic properties of the prepared samples were investigated.

2 Materials and Method

A self-made nanopowder was used as the raw material. The preparation process and properties of the powder are provided in reference [18]. The phase of the raw powder was $\text{Ni}_{0.75}\text{Zn}_{0.25}\text{Fe}_2\text{O}_4$ (NiZn ferrite, JCPDS No. 52-0277), and its average crystallite size of 16 nm was estimated by using the Scherrer equation [33]. The raw powder was granular, and its particle size ranged from 10 nm to 25 nm.

The sintered NiZn ferrite was prepared by using a pulse electric current sintering system (SPS-20T-10, Chenhua Co., Ltd., China). A schematic of the graphite die-punches setup that was used in the sintering system is shown in Fig. 1. The raw powder was placed into the graphite cavity that formed between the graphite die and punches. An axial pressure of 60 MPa was applied to the powder in the graphite cavity for 1 min. Then, the pressure was removed and a pulse direct current was applied to the die-punches-sample system. The sample was heated at a heating rate from 5°C/s to 25°C/s. When the sample temperature reached the preset sintering temperature that ranged from 900°C to 1200°C, the power supply was turned off, followed by a rapid cooling of the sample. A disc-shaped sintered sample with a diameter of 20 mm and a thickness of ~3 mm was obtained. For comparison, a traditional non-pressure solid-phase sintering method was used to prepare the sintered samples with the same raw powder and prepressing conditions. The sintering process diagrams of the NiZn ferrite that was prepared by the UPECS method and the traditional method are shown in Fig. 2.

Phase analysis of the raw powder and the prepared ceramics at the center of the disc-shaped samples was obtained by XRD (X'Pert PRO, PANalytic, Netherlands) using $\text{CuK}\alpha$ radiation ($\lambda = 1.5406\text{\AA}$) under the same XRD measurement conditions. The microstructures of the raw powder and ferrite ceramics were examined by field emission scanning electron microscopy (Zeiss Ultra 55, Germany). The average grain size of the sintered sample was estimated by a line intercept method on an SEM image. The static magnetic properties at room temperature and the Curie temperature of the samples were measured by using a vibrating sample magnetometer (Lake Shore 8607, America). The bulk density (D_b) of the ferrite ceramics was calculated using a water boiling method based on

Archimedes principle. The relative density (D_r) and porosity (P) of the sintered samples were calculated from equations (1) and (2), respectively:

$$D_r = \frac{D_b}{D_x} \times 100\% \quad (1) \quad \text{and} \quad P = \left(1 - \frac{D_b}{D_x}\right) \times 100\% \quad (2)$$

where D_x is the X-ray density.

3 Results and discussion

3.1 Phase crystal structure of samples

Figure 3a and b shows the XRD patterns of the samples that were sintered by the UPECS method at a fixed heating rate of 25°C/s while changing the sintering temperature, and at the fixed sintering temperature of 900°C while changing the heating rate, respectively. The diffraction peaks of all sintered samples were consistent with the standard diffraction peaks of the cubic spinel NiZn ferrite ($\text{Ni}_{0.75}\text{Zn}_{0.25}\text{Fe}_2\text{O}_4$, JCPDS No. 52-0277). These peaks corresponded to the (111), (022), (113), (222), (004), (224), (333), and (044) planes of the NiZn ferrite. The diffraction peaks of the sintered NiZn ferrites were stronger and sharper than those of the raw powder because of the improved crystallization that was induced through high-temperature sintering, even though the sintering time was short (no holding time).

The precise lattice parameter (a) of the sample was obtained by extrapolation. Equation (3) can be derived from the Bragg equation and the expression of interplanar spacing in the cubic crystal:

$$a = \frac{\lambda \sqrt{H^2 + K^2 + L^2}}{2 \sin \theta} \quad (3)$$

where λ is the wavelength of the incident X-ray in the XRD experiment; θ is the Bragg angle; H, K and L are the indices of the interference plane. The Nelson-Riley function $f(\theta)$ is used as the extrapolation function, which is expressed as [34]:

$$f(\theta) = \left(\frac{\cos^2 \theta}{\sin \theta} + \frac{\cos^2 \theta}{\theta} \right) / 2 \quad (4)$$

According to the XRD data of a sample, the lattice parameter values and $f(\theta)$ of the sample were calculated from equations (3) and (4), respectively, and the a - $f(\theta)$ plot was constructed. The fitting linear a - $f(\theta)$ plot was extrapolated to $f(\theta) = 0$ (i.e., $\theta = 90^\circ$) and the intercept was the precise lattice

parameter. The precise lattice parameter values of the samples are shown in Table 1. The difference in lattice parameters between the prepared and standard samples ($\text{Ni}_{0.75}\text{Zn}_{0.25}\text{Fe}_2\text{O}_4$, JCPDS No. 52-0277: $a = 8.370 \text{ \AA}$) was less than 0.05%. The result indicated that the main phase of the sintered samples that was prepared by UPECS was $\text{Ni}_{0.75}\text{Zn}_{0.25}\text{Fe}_2\text{O}_4$.

3.2 Microstructure of samples prepared by UPECS

3.2.1 Microstructure of samples prepared by UPECS at different sintering temperatures

Figure 4 shows the SEM images of the NiZn ferrite sintered by UPECS at different temperatures when the heating rate was 25°C/s . The SEM images of the sintered samples at a depth of $500 \mu\text{m}$ from the sample surface (marked as an edge) are shown in Fig. 4a–e and that of the center position (marked as the center) are shown in Fig. 4f–j. For the edge and center of the sample, the grain size of the NiZn ferrite increased gradually with the increase in sintering temperature from 900°C to 1200°C . The average grain size, density and porosity of the sample that was sintered by the UPECS are shown in Table 1.

When the sintering temperature was 900°C , the prepared sample was loose and the grains were small but nonuniform. The sample was composed of grain clusters, in which the grains were in contact with each other closely. Interconnected pores appeared between clusters (Fig. 4a and f), which resulted in a high sample porosity and low density. As the temperature increased to 1000°C , the grains combined closely and the pores were reduced. However, the difference in grain size remained obvious (Fig. 4b and g). When the sintering temperature reached 1050°C , the porosity decreased further, the grain shape was regular and the grains gradually tended to be uniform (Fig. 4c and h).

When the sintering temperature increased to 1100°C , the grains of the sample became more uniform and the grain size increased further. The pore size at the junction of the grains increased (Fig. 4d and i), and round pores appeared on the grain surface (Fig. 4i). Many pores in the sample originated from the partial reduction of Fe^{3+} and Zn volatilization. At a low oxygen partial pressure and high temperature of 1100°C , part of the Fe^{3+} was reduced, which caused partial NiZn ferrite decomposition to form O_2 and ZnO (equation 5). The low oxygen partial pressure and high temperature environment were conducive to ZnO decomposition into Zn and O_2 , and Zn

volatilization occurred because of its low boiling point of 907°C (equation 6).

The O₂ and Zn vapor that were produced in the reactions increased and/or expanded the pores in the sample, which resulted in the low density and high porosity of the sintered sample (Table 1). Although part of the NiZn ferrite decomposed at a high temperature and a low oxygen partial pressure, the NiZn ferrite decomposition was minimal because of the short UPECS sintering time and so the secondary phase was not detected by XRD. Therefore, the XRD patterns of the sintered samples indicated that the NiZn ferrite was the main phase (Fig. 3).

When the sintering temperature reached 1200°C, the sample was dense and most of the sample grains were larger than 1 μm (Fig. 4e and j). The pores in the sample were large and they mainly converged at the junction of the multiple grains. The grain size (D) increased with the increase in sintering temperature, which was closely related to the sintering time (t) and temperature (T). The relationship between D, t and T conformed to the crystal growth kinetics, as shown in equation (7) [35-37].

$$D^n - D^0 = t \cdot K_0 \cdot \exp(-Q/RT) \quad (7)$$

$$D^n \approx t \cdot K_0 \cdot \exp(-Q/RT) \quad (8)$$

where n is a grain growth exponent, D⁰ is the grain size before sintering, Q is the activation energy of the grain growth, and R is the gas constant. Because the D⁰ was very small and was ignored, equation (7) was simplified to equation (8). For similar reaction times, the energy in the sample increased with an increase in temperature and the grain boundary moved to reduce the interfacial energy and meet the minimum energy requirement. So, grains easily grew into large grains at a high temperature. Most pores in the sintered sample contained gases (O₂ and Zn vapor originated from part of the NiZn ferrite decomposition). The pressure in the small pores of the sample was higher than that of the big pores, which made small pores migrate to the large pores. During sintering, some pores were excluded from the sample through grain boundary diffusion, whereas other pores were not eliminated in time because of the short sintering time, and they converged at the junctions of the multiple grains. Therefore, with an increase in grain size, the pore size increased.

3.2.2 Microstructure of samples prepared by UPECS at different heating rates

Figure 5 shows the SEM images of the samples that were sintered at 900°C through UPECS with different heating rates. SEM images of the edge of the samples are shown in Fig. 5a–c and those of the center of the samples are shown in Fig. 5d–f. The average grain size, density and porosity of the sintered samples are shown in Table 1. For the edge and center of the sample, the grain size of the NiZn ferrite decreased and the porosity increased with an increase in heating rate from 5°C/s to 25°C/s. The average grain size of the sample sintered at 5°C/s ranged from 420 nm to 460 nm, and the sample was dense and homogeneous (Fig. 5a and d). The sample that was sintered at 10°C/s remained dense and its average grain size decreased slightly to 398–410 nm, but the grains became uneven (Fig. 5b and e). As the heating rate increased to 25°C/s, the sintered sample was porous and its grain size distribution was wide (Fig. 5c and f). The large grain exceeded 600 nm and the small grain was less than 100 nm.

The above experimental results indicated that the NiZn ferrite samples were kept above 600°C for no more than 90 s during UPECS. The crystallization of the prepared sample was better than that of the raw powder, and the highest relative density of the sintered sample reached 95.72%. Compared with the traditional sintering method with a high temperature for several hours [10–12], the rapid sample densification by UPECS was attributed to the heating mechanism in the pulse current sintering. The grain at the edge of the sample was larger than that at the center of the sample, which was related to the pulse current heating approach and the conductivity characteristics of the NiZn ferrite.

3.3 Discussion on UPECS mechanism and microstructure inhomogeneity of NiZn ferrite

3.3.1 Discussion on the UPECS mechanism of NiZn ferrite

A high current and low voltage is used in pulse current sintering, which can reduce the sintering time significantly because of the heating mechanism. Compared with the external heat source in the traditional sintering, the energy to heat the sample originates from Joule heat conduction from the graphite die-punches setup and the Joule heat that is generated by the NiZn ferrite during pulse current sintering. NiZn ferrite is a semiconductor material. Its conductivity (σ) correlates with the Arrhenius equation, which is expressed as [38]:

$$\sigma = A \exp (-\Delta E/2kT) \quad (9)$$

where σ_0 is a constant that is related to the carrier concentration, ΔE is the conductivity activation energy, k is the Boltzmann constant and T is the absolute temperature. According to equation (9), the NiZn ferrite conductivity increases with an increase in temperature.

Figure 6 shows a schematic diagram of the current flow path during sintering. Compared with graphite, NiZn ferrite was considered to be an insulator because of the low temperature and low density at the initial stage of sintering. The current deflected at the front end of the upper punch and flowed through the graphite die rather than directly through the sample in the cavity along the axial direction (Fig. 6a). The energy that heated the sample originated almost exclusively from the heat conduction of the Joule heat that was generated in the graphite die-punches setup at the initial sintering stage. Progress in sintering resulted in an increase in NiZn ferrite conductivity with an increase in temperature. The conductivity also increased with the increasing sample density. Therefore, part of the current flowed through the sample (Fig. 6b) and the sample itself generated Joule heat in the middle and final sintering stages. The sample temperature was elevated rapidly because of the synergistic effect of heating energy concentration of the current and the conductivity characteristic of the semiconductor NiZn ferrite.

During pulse current sintering, the Joule heat that was generated at the contact point between particles was important for NiZn ferrite grain growth and densification. Some particles contacted each other in the green compact. When the sample temperature was high enough, part of the current flowed through the particles that contacted each other (Fig. 7a) and a large amount of Joule heat was generated at the contact points, which made the particle surface layer melt immediately to form necks instantaneously. As the current continued to flow through the sample, a large amount of Joule heat was still generated at the necks, which increased the atom/molecule diffusion at the neck and promoted grain growth and densification of the sample. The Laplace stress (marked as P, Fig. 7b) appeared along with neck formation. The Laplace stress was a tensile stress for the necks, which increased the interaction between particles and results in the promotion of neck growth. However, the Laplace stress was a compressive stress for the pores near the necks, which caused the pores to shrink and led to rapid sample densification. The above results showed that a large amount of Joule

heat and Laplace stress at the necks promoted NiZn ferrite densification and grain growth during pulse current sintering, which completed the NiZn ferrite sample sintering within tens of seconds.

3.3.2 Discussion on the microstructure inhomogeneity of NiZn ferrite prepared by UPECS

Figure 4 shows that when the heating rate was high (25°C/s), the grains at the edge of the sample that was obtained by the UPECS method were significantly larger than those at the center of the sample. The difference in grain size of the sample is shown in Fig. 8a and Table 1. The grain inhomogeneity in the sample originated from the uneven temperature distribution in the sample during sintering. In the early stage of UPECS, the heating energy for the sample originated only from the heat conduction of Joule heat that was generated in the graphite setup. The time that the sample was at a high temperature was short because the sample was heated to the sintering temperature fairly rapidly, followed by a turning off of the supply power and rapid sample cooling. During this short heat treatment, the temperature distribution in the sample was uneven at the early sintering stage. The temperature at the edge of the sample, which was in direct contact with the graphite setup, was higher than that at the center of the sample. The uneven temperature distribution in the sample led to an uneven current distribution in the sample in the middle and final sintering stages because of the characteristic of a negative temperature coefficient of resistivity of the NiZn ferrite (Fig. 6b). The Joule heat energy that was generated at different positions in the sample self was different, which aggravated the temperature difference between the center and edge of the sample. Generally, a high temperature promoted grain growth, so the grains at the edge of the sample were larger than those at the center of the sample because of the uneven temperature distribution in the sample. Although the temperature in the sample may be homogeneous because of heat conduction during the final sintering stage, the difference in grain size distribution that was caused during the earlier sintering stage was partially retained in the final sintered sample because of the short sintering time.

Figure 4, especially Fig. 4d and i shows that more pores existed at the center of the sample compared with those at the edge of the sample. The temperature at the edge of the sample was higher than that at the center of the sample, which yielded a better migration ability of the pores in the former. The pores at the edge of the sample were removed more easily from the sample by grain

boundary diffusion than those at the center of the sample. Based on the above two reasons, fewer pores existed at the edge of the sample (Fig. 4d) compared with those at the center of the sample (Fig. 4i).

3.3.3 Comparison of microstructure between samples sintered by different methods

By using the same raw powder, prepressing conditions and at the sintering temperatures of 900°C and 1050°C, NiZn ferrite samples were prepared by the UPECS method at a heating rate of 25°C/s and by the traditional sintering method at a heating rate of 10°C/min, respectively. The sintering process diagrams are shown in Fig. 2. Figures 9 and 10 show the XRD patterns and SEM images of the center of the sintered samples, respectively. Figure 9 shows that the main phase of the samples sintered by the two methods was the same, which was the NiZn ferrite ($\text{Ni}_{0.75}\text{Zn}_{0.25}\text{Fe}_2\text{O}_4$, JCPDS No. 52-0277). The crystallization of the sintered samples was better than that of the raw powder. Figure 10 shows that the samples that were sintered at 900°C were loose and the samples that were sintered at 1050°C were dense for both samples that were sintered by the two methods. The average grain size of the NiZn ferrite that was prepared by the UPECS method was slightly smaller than that of the NiZn ferrite that was prepared by the traditional sintering (Table 1) because of the short UPECS time. The experiment indicated that the NiZn ferrite with a similar density and crystallization was prepared within tens of seconds by the UPECS method, whereas traditional sintering requires several hours, which confirmed that NiZn ferrite densification and grain growth was accelerated by the UPECS technique.

3.4 Magnetic properties of sintered samples prepared by UPECS

Hysteresis loops of the raw powder and NiZn ferrite at the center of samples that were prepared by UPECS are shown in Fig. 11. These narrow loops exhibited characteristics that were typical of soft magnetic materials. For example, the coercivity (H_c) and residual magnetization (M_r) were small and the specific saturation magnetization (M_s) was high (Fig. 11 and Table 1). M_s is the intrinsic property of the magnetic materials, which is mainly related to its composition, crystallinity and particle size. The M_s of the sintered sample was higher than that of the raw nanopowder. This behavior was attributed to the good crystallization of the sintered sample, which avoided/reduced the decrease in magnetic moment that was caused by the spin disorder on the nanoparticle surface.

The M_s of the sintered samples fluctuated slightly, which was attributed to a small difference in phase composition of the sintered samples because of the decomposition of a small amount of NiZn ferrite (equation 5) and Zn volatilization (equation 6) during the sintering.

The average grain size (D) dependence of H_c at the center of the samples that were prepared by the UPECS method is shown in Fig. 12. When the grain size of the sample increased from 294 nm to 1463 nm, the H_c of the sample increased first and then decreased. The sample H_c reached the maximum value of 36.0 Oe when the average grain size was 450 nm. According to the Stoner-Wohlfarth mono-domain theory [39], the magnetocrystalline anisotropy energy of the mono-domain grain increases with an increase in grain size, which occurs mainly because of the improvement in magnetocrystalline anisotropy. When the grain size increases to a critical value (D_c) between the dual- and mono-domain states, H_c reaches a maximum. The domain structure changes from the mono-domain state to the dual- or multi-domain state as the grain size exceeds D_c . Accordingly, the magnetization mechanism changes from domain rotation to domain wall movement and the H_c decreases with an increase in grain size [40]. Therefore, we inferred that the D_c between dual- and mono-domain states of the prepared NiZn ferrite was ~ 450 nm.

The Curie temperatures (T_c) at the center of the prepared samples are shown in Table 1. The T_c of the sintered samples varied from 503°C to 522°C, which showed only a small difference. T_c is one of the intrinsic parameters of ferromagnetic materials. It is only related to the chemical composition and crystal structure of the materials, and is almost independent of grain size, orientation, and stress distribution. The main phase of all sintered samples was the NiZn ferrite with good crystallization, so the T_c of the sintered samples was similar. The slight difference in sample T_c was mainly attributed to the small fluctuation in phase composition that was caused by the decomposition of a small amount of NiZn ferrite and Zn volatilization during sintering. The T_c of the sintered sample was lower than that of nickel ferrite (587°C) [41], which was attributed to the nonmagnetic Zn^{2+} in the NiZn ferrite. The nonmagnetic Zn^{2+} ions that occupied A-sites in the NiZn ferrite reduced the superexchange between the A–B sublattices, which resulted in a decrease in sample T_c .

4 Conclusions

The dense NiZn ferrite ceramics were prepared using the UPECS method, during which the sample was kept above 600°C for no more than 90 s. The conclusions obtained from this work were as follows:

(1) Grain growth and densification of the NiZn ferrite were accelerated, which benefited from the synergistic effect of the energy concentration of pulse current and joule heating of the sample.

(2) The difference in grain size between the edge and center of the samples that were prepared by the UPECS method originated from the uneven temperature distribution and current distribution in a sintering sample. The low heating rate and sintering temperature favored NiZn ferrite formation with a high density and ultrafine grains by the UPECS method.

(3) The slight differences in M_s and T_c of the prepared samples were mainly attributed to the small fluctuation of phase composition that was caused by the decomposition of a small amount of NiZn ferrite and Zn volatilization during sintering. The nonmagnetic Zn^{2+} ions that occupied A-sites reduced the superexchange interaction between A- and B-sublattices, which decreased the T_c of the prepared sample.

(4) According to the grain size dependence of H_c , it was inferred that the critical grain size between the dual- and mono-domain states of the sintered NiZn ferrite may be ~450 nm.

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Figure captions

- Figure 1** Schematic of a graphite die-punches setup used in the UPECS method.
- Figure 2** Sintering process diagrams of NiZn ferrite using (a) UPECS and (b) traditional sintering
- Figure 3** XRD patterns of samples prepared by UPECS at different (a) sintering temperatures and (b) heating rates.
- Figure 4** SEM images of (a)–(e) the edge and (f)–(j) the center of samples sintered at different temperatures through UPECS.
- Figure 5** SEM images showing (a)–(c) edge and (d)–(f) center of samples sintered at different heating rates through UPECS.
- Figure 6** Schematic diagram of current flow path at (a) early, and (b) intermediate and final stages of sintering.
- Figure 7** Schematic diagram of (a) current flow through particles and (b) “neck” formation between particles on the base of Joule heating.
- Figure 8** Average grain size of samples sintered at different (a) temperatures and (b) heating rates by UPECS.
- Figure 9** XRD patterns of samples sintered by UPECS and traditional method.
- Figure 10** SEM images showing center of samples sintered by (a), (b) UPECS and (c), (d) traditional method.
- Figure 11** Hysteresis loops of samples prepared at different (a) sintering temperatures and (b) different heating rates by UPECS.
- Figure 12** Grain size dependence of coercivity of prepared ceramics.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Table 1 Sintering conditions, lattice constant, average grain size, density, porosity and magnetic properties of NiZn ferrites

Sintering method	Molding pressure (MPa)/dwell time (min)	Heating rate	Sintering temperature (°C)	Duration time not less than 600°C	Lattice constant (Å)	Average grain size (nm)		Density (g/cm ³)	Porosity (%)	Magnetic properties			
						Edge	Center			M _s (emu/g)	M _r (emu/g)	H _c (Oe)	T _c (°C)
Raw powder	--	--	--	--	8.366	--	--	--	--	43.9	0.2	0.2	498
UPECS	60/1	25 °C/s	900	42 s	8.370	337	294	4.49	16.06	61.6	2.2	21.9	522
UPECS	60/1	25 °C/s	1000	56 s	8.366	533	435	4.78	10.64	61.3	1.3	32.6	513
UPECS	60/1	25 °C/s	1050	63 s	8.370	621	450	4.86	9.14	63.0	2.1	36.0	505
UPECS	60/1	25 °C/s	1100	70 s	8.369	1018	754	4.71	11.95	60.6	1.2	24.4	511
UPECS	60/1	25 °C/s	1200	84 s	8.367	1787	1463	5.12	4.28	60.1	1.2	23.3	503
UPECS	60/1	5 °C/s	900	90 s	8.370	456	421	5.11	4.47	62.9	3.8	27.7	503
UPECS	60/1	10 °C/s	900	60 s	8.370	410	398	5.09	4.84	62.8	1.3	25.4	504
UPECS	60/1	25 °C/s	900	42 s	8.370	337	294	4.49	16.06	61.6	2.2	21.9	522
Traditional method	60/1	10°C/min	900	300 min	8.367	--	225	4.85	9.33	62.4	1.5	19.8	505
Traditional method	60/1	10°C/min	1050	315 min	8.368	--	750	5.01	6.34	64.0	1.1	24.9	512

* Average grain size was estimated by a line intercept method on SEM image.

Figures

Figure 1

Schematic of a graphite die-punches setup used in the UPECS method.

Figure 2

Sintering process diagrams of NiZn ferrite using (a) UPECS and (b) traditional sintering

Figure 3

XRD patterns of samples prepared by UPECS at different (a) sintering temperatures and (b) heating rates.

Figure 4

SEM images of (a)–(e) the edge and (f)–(j) the center of samples sintered at different temperatures through UPECS.

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Schematic diagram of current flow path at (a) early, and (b) intermediate and final stages of sintering.

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SEM images showing center of samples sintered by (a), (b) UPECS and (c), (d) traditional method.

Figure 11

Hysteresis loops of samples prepared at different (a) sintering temperatures and (b) different heating rates by UPECS.

Figure 12

Grain size dependence of coercivity of prepared ceramics.