New Magnetic Composite Based on Ni-Zn for Magnetic Screenings and Power Conversion with Its Recyclable and Formulable Features

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Abstract-The development of ferrites makes them can be widely used in the field of optics, electrics, sound, magnetism. Ni-Zn ferrite offer the possibilities of good magnetic properties and high electrical resistivity, together with large permeability at high frequency. However, the present Ni-Zn ferrite's properties are strongly influenced by the materials' composition and microstructure, which are sensitive to the preparation methodology used in their synthesis. Furthermore, it also need ferrite to answer strict environment and realize better properties with the recent trend. So, more improvement should be achieved. Here we proposed a modified way to prepare it, new polymer bonded Ni-Zn ferrite. We make effort to improve its magnetic property and formulate its tendency. We demonstrated to optimize the preparation technology including modifying the method of preparing magnetic powder and bonding with binder. Selection of materials is also important to its property. Through this work, new polymer bonded Ni-Zn ferrite presents high magnetic property and recyclable property. Further, through large amount of experiment data and basic knowledge, we can also formulate its magnetic property corresponding to its ratio.

Keywords-Polymer, ferrite, preparation, recyclable, formulate.

I. Introduction

Ni–Zn ferrites are soft ferrimagnetic materials having high resistivity values and low magnetic coercivity [1] and large permeability at high frequency [2]. Owing to good magnetic properties and high electrical resistivity, Ni-Zn ferrite could be an excellent material for power transformers in electronics, storage devices, microwave devices, magnetic fluids, and telecommunication applications, etc. [3–5].

However, the present ferrites are prepared by the conventional ceramic method, in which the stoichiometric composition and microstructure are extremely difficult to control. Additionally, the properties of Ni-Zn ferrite materials are strongly influenced by the materials' microstructure and composition, which are sensitive to the preparation methodology, such as the sintering conditions and the impurity levels present in or added to these materials could change their properties [6]. Therefore, the standardization and selection of an appropriate process is, the key to obtain good quality ferrites. The conventional method requires prolonged heating at high temperatures during preparation, which may cause some of the constituents to evaporate, thereby modifying the desired stoichiometry [7]. Moreover, in Ni–Zn ferrite, the volatilization of zinc at high temperatures results in the formation of Fe^{2+} ions, which increase electron hopping and reduce resistivity. The grinding or milling operations involved in the process lead to the loss of some material and to concentrations of impurities that result in non-stoichiometric compositions [8,9]. Consequently, samples always present unreproducible characteristics. So, we proposed a new way to bypass this problem.

Here we choose polymer bonded methods as a better alternative, since they overcome the instability of the conventional ceramic method, together with making the most of its advantage including efficiency and safety. The polymer bonded method has proved to be one of the effective routes because of its controllability and high efficiency. Including to decrease the cost and waste of preparation thus increase its efficiency. Varieties of bonded methods such as co-precipitation, hydrothermal synthesis, the citrate precursor method, the glass-ceramic route and the sol–gel process have been developed [10-12].

With the recent trend toward technology efficiency and excellence, polymer bonded ferrite must realize high magnetic property and mechanical property in an effective way. For this purpose, we make effort to optimize technology and materials, aim for getting high performance polymer bonded ferrite.

In this study, we prepared polymer-bonded ferrite through bonding magnetic powders and binder such as thermoplastic polymer at an appropriate mixing ratio with different methods by compression molding. The magnetic powder was usually obtained by gas atomization, melt quenching. Here we propose 2 methods to prepare magnetic powder through combustion reaction or the method that sintering and then smash it through atomization and ball milling. The combustion reaction synthesis method of magnetic powder preparation is a good choice, since they not only overcome the drawbacks of the conventional ceramic method, but also prepare nano-scale magnetic powders possess distinctive physical and chemical properties because of their nano-sized crystallite, large surface area and different surface properties (such as surface defect) etc. [13]. Then bonding magnetic powder and binder, mixture was then molded by machine. The advantage of this comprehensive method is that we can not only get good performance raw magnetic powder, but also can get the optimal powder size which is so important to the binding efficiency. Additionally, this method has the advantages of applying inexpensive raw materials and maintaining a relatively easy preparation process, moreover achieving a fine magnetic powder with high homogeneity. Hence, we can improve efficiency and magnetic performance through this new method.

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The method mixing magnetic powder and binder is also important to its property. The magnetic powder was first coupled with the acetone solution containing a silane coupling agent for improving bonding effect because magnetic powder is hydrophilicity and binder is hydrophobicity, through couple treatment, they can form strong chemical bonds. The schematics of coupling agent is shown in Fig. 1. Thus, it will increase the mechanical property of polymer bonded ferrite.

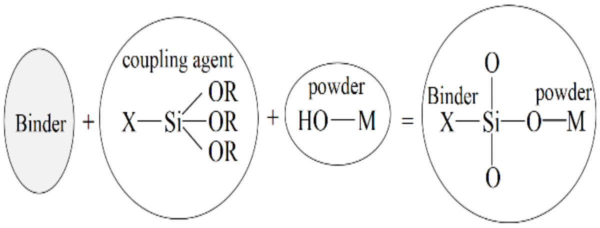

Fig. 1: Schematics of coupling agent.

In this study, we also optimize preparation process through improving the powder preparation and controlling its contents. We bypass the obstacle by calculating the magnetics parameters and clarify the relationship between the preparation conditions (magnetic powders and particle size of starting materials) and some properties of the bonded ferrite. We also design a recyclable polymer bonded ferrite through materials and process design under certain conditions.

II. Experimental procedure

The flow chart of the experimental procedure is shown in Fig. 2.

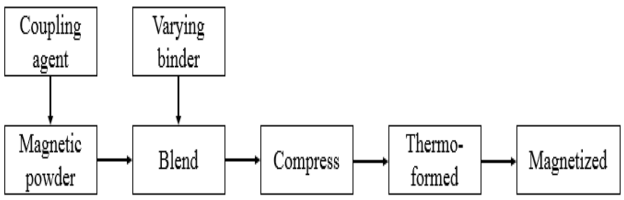

Fig. 2: Flow chart of experimental procedure in this study.

The samples were prepared by mixing magnetic powder and binder, and then combined through a compression-molding technique.

Here we propose 2 methods to prepare magnetic powder: sintering and combustion reaction.

The combustion reaction synthesis method, mix oxidizing reagents (such as nitrates) and reducing reagent (such as urea). And then the reaction happens spontaneously, this is because it is an exothermal reaction, thus, high temperatures generated will make oxides crystallize easily. Additionally, the reaction also prevents the instantaneous formation of particle agglomeration which will decrease product property dramatically [14]. The particle produced by this process have the advantage of chemical homogeneity on an atomic scale, uniform particle sizes and controlled particle shapes. Moreover, the reaction will not cause the loss of materials and impurity because there is no other media or steps. It can form magnetic powder directly [15-17]. This fact is important because contrast to the microstructures developed during indirect method (many steps to prepare magnetic powder), to a large extent, its powder's characteristics (crystallite size and shape, size distribution, porosity, state of agglomeration, chemical and phase composition) is more efficient and helpful to prepare magnets [18], which are closely associated with the processing method. Several investigations of the characteristics of Ni–Zn ferrites, such as their structure, electrical conductivity, chemical and elastic properties, dielectric behavior and magnetic properties, have been reported in the literature [19-21]. However, no study about polymer bonded magnets use Ni–Zn ferrite powders produced by this combustion reaction method. This study, investigated the character, the effect of the contents on the preparation efficiency and magnetic properties prepared by combustion reaction, then prepared with comprehensive method next step.

Combustion reaction method, showed in Fig. 3. Magnetic powder with a nominal composition of $Ni_{(1-x)}Zn_xFe_2O_4$, where x= 0.3, 0.4 and 0.5was prepared, including a heating reaction $(NiO+ZnO+Fe_2O_3=Ni_a Zn_{(1-a)} Fe_2O_4)$ of a series mixture of metallic oxide (Ni, Zn and Fe), an exothermic reaction of a mixture of metallic nitrates (Ni, Zn and Fe) and urea (reducing agent). The materials used were zinc nitrate $Zn(NO_3)_2{\cdot}6H_2O$, nickel nitrate $Ni(NO_3)_2{\cdot}6H_2O$, iron nitrate $Fe(NO_3)_3{\cdot}9H_2O$ and urea (NH_2CONH_2). The proportions of the initial reagents were calculated based on the total valences of the reacting elements to maximize magnetic property and preparation efficiency, following the concepts of physics and chemistry [22-23]. Equation as follows:

$$(0.5-x)Ni(NO_3)_2\cdot 6H_2O + 0.5 - xZn(NO_3)_2\cdot 6H_2O + 2Fe(NO_3)_3\cdot 9H_2O + 8NaOH = Ni_{(0.5-x)}Zn_{(0.5-x)}Fe_2O_4 + 8NaNO_3 + 28H_2O.$$ [24]

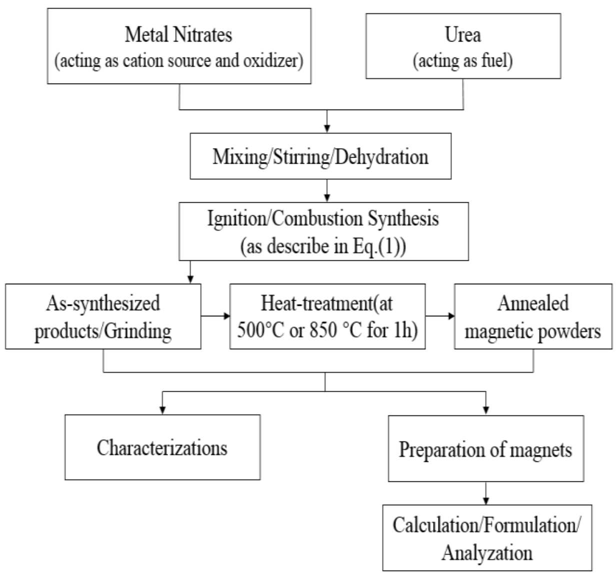

Fig. 3: Flow chart of combustion reaction method in this study

Sintering method, powder Fe_2O_3, NiO, CuO, ZnO were chosen as the raw materials to prepare NiZn ferrites. The molar percentage of the raw materials is listed in Table 1. First, all oxides were mixed with alcohol in a ball mill, according to their molar ratios. The mixture was then milled to 0.8 mm average particle sizes, respectively. Then the slurry was dried at 70°C. Second, pre-sintering them in muffle furnace at 950°C for 3h. The calcinations were then conducted in air for 3h. Third, the calcined powders were mixed with distilled water and 0.2mol% V_2O_5 then ground in a centrifugal ball mill until the powders reached an average particle size of 0.8 mm. The powders were then dried at 70°C. Fourth, calcine them in muffle furnace at 1150°C for 3h. The calcinations were then conducted in air for 3h. Finally, ball mill them into appropriate particle size for next step.

Table 1: Chemical compositions of samples

Sample no.	Fe_2O_3 (mol%)	ZnO (mol%)	NiO (mol%)	CuO (mol%)
1	50	30	20	0
2	50	30	17.5	2.5
3	50	30	15	5

The magnetic powder was then coupled with the acetone solution containing a silane coupling agent and was dried in an oven. The coupled magnetic powder was mixed uniformly with varying binder proportions of PMMA and curing agent [25]. The resulting mixtures were compressed to E-core samples at different pressure levels. Then thermoformed for two hours at different thermosetting temperatures. The samples were then magnetized using a high pulse magnetic field magnetizer at a voltage of 1400 V, which was sufficient to generate 4500 Oersted field strength to saturate the bonded Ni-Zn ferrites. The magnetic properties were measured by a HIOKI3532-50 LCR HiTESTER. All tests were carried out at room temperature.

III. Results and discussion

III.I. Magnetic properties

A series of preparation process were needed to prepare our sample in different conditions to determine the magnetic property and mechnical property. We label method 1 as preparing magnetic powders by combustion reaction and then bonded with epoxy resin, method 2 as preparing magnetic powders by sinstering method and then bonded with epoxy resin.

Fig. 4 shows the morphology of magnetic powders produced by 2 methods. From this picture, we can analyze powder's characteristics (powder size, degree of purity) visually. It shows that the powders prepared by method 1 , its powders size is smaller and powder is more pure compared with powder prepared by method 2. The light spot shows impurities exist in powders prepared by sinstering ways, it is because when ball milling it, some impurities doped.

Fig. 4: Morphology of magnetic powders prepared by 2 methods. method1(on the right), method2(on the left)

Fig. 5 presents the morphology of polymer bonded ferrite with different preparation, we can see that E-core prepared by method 1(on the right) is more uniform. Additionally, when we test their mechanical property, sample prepared by method 2 is more brittle and easily destroyed once we exert big force. Its mechanical property is worse compared with samples prepared by method 1.

Fig. 5: Morphology of E-core prepared by 2 methods method1(on the right), method2(on the left)

From this we can see that powder property prepared by method 1 is superior to method 2 whether in powder size or uniformity. Additionally, mechnical property is also corresponding to powder property.

III.II. Magnetic properties

Magnetic property determines whether polymer bonded ferrite can be commercially used or not. To get high-performance ferrite, we try to clarify the correlation between the material constants and μ_r in the polymer bonded ferrite which will help us optimize technology and formulate results.

The result μ were measured by a HIOKI3532-50 LCR HiTESTER. All test results below were calculated at the frequency of 50KHz.

The ratio of binder is an important factor that influences magnetic properties. Table 1 presents the magnetic properties of 1.5wt% - 6.0wt% binder ratio bonded magnets with same preparation technology. All experiment results have set standards and control variables, such as all materials should be the same, and prepared in 1 batch. Then calculate magnetic properties according to the following formulas:

$$\mu_e = \frac{L\,L_e}{0.4\pi N^2 A_e}$$

where L is sampler's length(cm); Le is the Inductance(μH), it is measured by HIOKI3532-50 LCR HiTESTER; N is the numbers of turns; A_e is effective sectional area(cm^2).

From this table, we can see that μ of the bonded ferrite decreased with the increasing of the ratio, while mechanical property increased when binder content increased. Moreover, when the ratio is less than 5%, the μ decreases slower than the ratio is bigger than 5%.

Table 1: Magnetic properties of magnets containing 1.5 wt% - 6.0 wt% binder content at 620 MPa pressure

Binder content	μ_e
1.5 wt.%	107.9
2.0 wt.%	104.4
2.5 wt.%	102.7
3.0 wt.%	100.0
3.5 wt.%	99.3
4.0 wt.%	96.8
4.5 wt.%	93.4
5.0 wt.%	92.5
5.5 wt.%	83.5
6.0 wt.%	80.2

Fig. 6 presents the tendency chart of different bonded ferrite with corresponding ratio, we can see variation tendency is not sharp, thus we can formulate tendency of μ through formula on the base of physics.

$$\mu = -0.05x^3 + 0.7x^2 - 4.7x + 111.8$$

x is the ratio of binder.

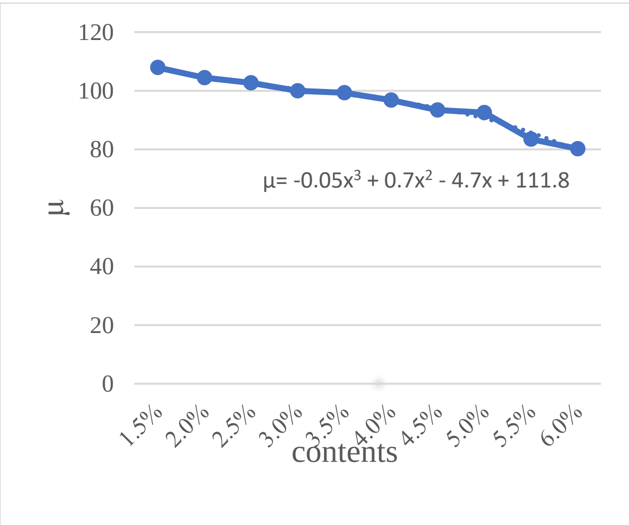

Fig. 6: Magnetic property of different ratio bonded ferrite.

The preparation method greatly determines its magnetic property. To get effective magnetic powders, we use 2 methods to prepare which were mentioned above, then addressed them with varying parameters.

Fig.7 presents the μ of samples with different parameters. It shows that the role of compacting pressure and binder's ratio is too small compared with catagolories of magnetic powder in influence magnetic property. The caculated μ prepared by method 1 is average 100, and few change whatever ratio and pressure changed. The same results can be indicated in method 2. It shows that category of magnetic powder is determining factor that influence magnetic property of bonded ferrite. Moreover, magnetic powder is determined by its preparation method. Method 1 is superior than method 2.

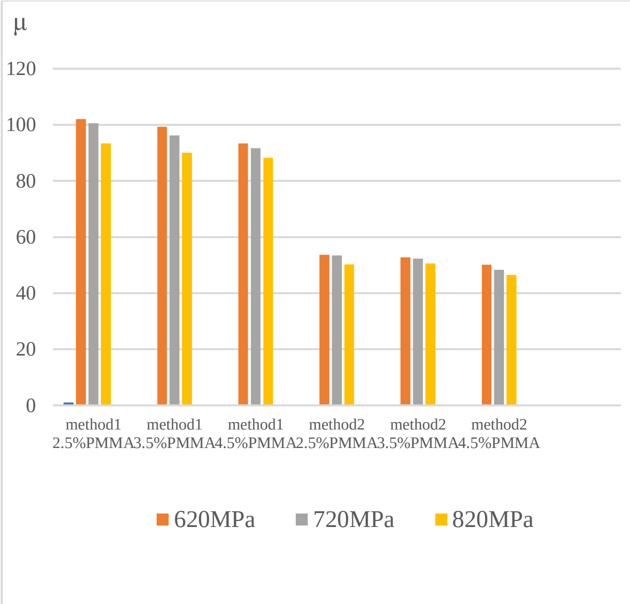

Fig. 7: Magnetic property prepared by 2 methods.

Different frequency may affect the u_r, it will influence application. So, we measured samples in different frequency, results showed in Fig.8. We can see in high frequency 50KHz or 100KHz, the μ are almost the same with each other, its magnetic property is so stable in high frequency.

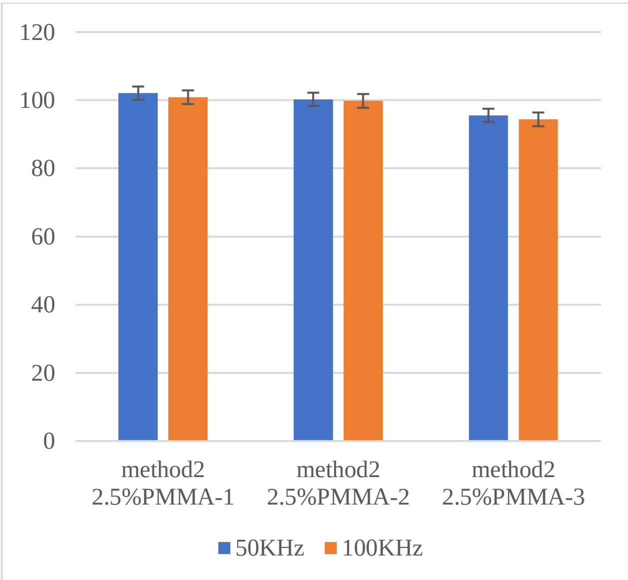

Fig. 8: Magnetic property measured in different frequency

III.III. Recyclable

Because when we prepare polymer bonded ferrite, some needed binder can be highly dissolved in aceton solution. So we try to design a new kind of ferrite in the investigation which was to develop recyclable polymer bonded ferrite with good performance at the same time. We designed a soluable binder which contain resin and curing agent, owing to they can not only thermoformed, but also dissolve to acetone under certain conditions. That means we can reuse it as a possible material for next time.

Fig. 9 presents the changes of recyclable ferrite after adding various amounts of acetone on the surface. The left one is that adding some drops of acetone, then press it. From the picture, we can see that dissolved area only exist in the areas where add acetone. The right one is that adding more acetone, then press it. We can see that the most of sample was dissolved. Thus, the recyclable polymer bonded ferrite can be dissolved in corresponding acetone. Such recyclable feature could be appealing in applications because it can save materials.

Fig. 9: changes of recyclable ferrite after a period of time

IV. CONCLUSIONS

The polymer bonded ferrites is efficiency and recyclable owing to the flexibility of materials and preparation technology. Further,we formulate the μ with different ratio based on physics.

We have developed a new polymer bonded ferrites using novel preparation technology with a series way of preparing magnetic powder, blending, and thermoformed. Different prepartion and different materials can infuence the magnetic property, meanwhile, compared with ratio and molding conditions, prepation of magnetic powder play a more improtant role. The magnetic powder prepared by combustion method is superior to the powder prepared by sinstering method whether in mechanical or magnetic property. We also note that the μ/ratio can be calculated on the basis of the measured, and physics. The magnetic property is also stable in high frequency. Such polymer bonded ferrites have the potential to be cost effective and recyclable, and to have applicable magnetic property.

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