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Study of AC magnetic properties and core losses of Fe/Fe_3O_4-epoxy resin soft magnetic composite

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Abstract

Soft Magnetic Composites (SMC) were prepared by coating of nanocrystalline Fe_3O_4 particles, synthesized by co-precipitation method, on atomized iron powder of particle size less than 53 µm in size using epoxy resin as a binder between iron and Fe_3O_4. Fe_3O_4 was chosen, for its high electric resistivity and suitable magnetic properties, to keep the coating layer magnetic and seek improvement to the magnetic properties of SMC. SEM images and XRD patterns were recorded in order to investigate the coatings on the surface of iron powder. A toroid was prepared by cold compaction of coated iron powder at 1050 MPa and subsequently cured at 150˚C for 1 hr in argon atmosphere. For comparison of properties, a toroid of uncoated iron powder was also compacted at 1050 MPa and annealed at 600˚C for 2 hr in argon atmosphere. The coated iron powder composite has a resistivity of greater than 200 µΩm, measured by four probe method. A comparison of Magnetic Hysteresis loops and core losses using B-H Loop tracer in the frequency range 0 to 1500 Hz on the coated and uncoated iron powder is reported.

Keywords: Soft Magnetic Composite, High frequency applications, Core losses

1 Introduction

Soft magnetic composites (SMC) are electrically insulated iron/iron alloy powders that may be used for high frequency electrical applications due to low core losses and 3D magnetic flux propagation capabilities [1-3]. Soft magnetic materials with high permeability and low losses are of interest for high frequency (400 Hz to few KHz) and high speed electric motors. Sintered soft iron

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powder compacts are used for DC applications, for low frequency (few hundred Hz) applications laminated steels are used [4, 5]. However, sintered iron compacts and laminated steel are not useful for high frequency applications due to eddy current losses when used in AC magnetic fields. Soft ferrites are used at high frequencies (few MHz to GHz) due to their high electrical resistivity. However ferrites possess lower saturation induction in comparison with soft iron containing alloys. Electrically insulated iron powders (SMC) with better magnetic properties and low core losses may hence be used for applications in the frequency range few kHz. The electrical insulation on the iron powder particles should lower the eddy current losses. The insulation layer characteristics need also be optimized for better permeability and core losses [6-9]. Electrical insulations based on inorganic compound makes it possible for high temperature heat treatments that may yield the better magnetic properties in the final product in comparison to organic compound based insulations. This paper presents a study of SMC prepared by insulating micrometer sized iron powder with uniform nano sized Fe_3O_4 particles and epoxy resin.

2 Experimental Details

2.1 Materials

Atomized iron powder of the grade JIP (KIP) 304A, procured from JFE Steel Corporation Tokyo, was first sieved and separated according to particle size. Iron powder with particle size of less than 53 µm (-270 ASTM mesh) was used for the composite preparation. For Fe_3O_4 preparation $FeCl_2.4H_2O$, $FeCl_3$ and NaOH pellets sourced from Merck India were used. Also for the Epoxy resin, Araldite® 506 epoxy resin and 4,4’-Methylenedianiline hardener supplied by Sigma Aldrich and Acros Organics were used.

2.2 Fe_3O_4 preparation by co-precipitation method

The stoichiometry of the chemicals was determined by using the following equation.

$$Fe^{2+} + 2Fe^{3+} + 8OH^{-} \rightarrow Fe_3O_4 + 4H_2O \quad (1)$$

The reaction solution was prepared by dissolving 4.5 millimol of $FeCl_2.4H_2O$ and 9 millimol of $FeCl_3$ in 100 ml distilled water followed by stirring at 80˚C under N_2 gas atmosphere. 36 millimole of NaOH were dissolved in 100 ml distilled water. The prepared NaOH solution was added drop by drop into the reaction solution and the reaction was performed at 80˚C under N_2 gas atmosphere, until black precipitate of Fe_3O_4 was formed. The solution was decanted and the precipitate of Fe_3O_4 washed several times using distilled water till chloride ions were removed. The prepared Fe_3O_4 particles were in the range of 20-50 nm and were characterized by powder X-ray diffraction (XRD).

2.3 Composite preparation

A composite was prepared with volume percentages of iron, Fe_3O_4 and epoxy resin in the ratio of 95, 2.5 and 2.5 respectively. First Araldite® 506 epoxy resin and 4, 4’-Methylenedianiline were dissolved in acetone. 4,4’-Methylenedianiline is the hardener for the epoxy resin and the resin to hardener ratio was maintained at 4:1 by weight. Fe_3O_4 nano powder was added into this followed by stirring for uniform distribution of Fe_3O_4 nano particles and epoxy resin. Now the iron powder was added to this mixture and stirred until all of the acetone evaporated. This resulted in the coating of

Fe_3O_4 and epoxy resin on the iron powder. The coated iron powder was then dried and cured at 70ºC in air.

A toroid of dimension outer diameter = 35 mm, inner diameter = 25 mm was compacted at 1050 MPa using the composite powder mixture. This toroid was post cured at 150˚C in argon atmosphere for 2 hr. The coating on the iron powder was characterized by powder X-ray diffraction (XRD) and scanning electron microscope imaging (SEM) techniques. For comparison of properties uncoated iron powder was also compacted at 1050 MPa as a toroid of similar dimensions and annealed at 600ºC for 2 hr in argon atmosphere.

Toroids were wound with primary (400 turns) and secondary (300 turns) copper windings for measurement of AC magnetic properties and core losses using a hysteresis loop tracer in the frequency range 0-1500 Hz, made by Laboratorio Elettrofisico Walker LDJ Scientific, Italy. Resistivity of the toroids was measured by the four probe method.

3 Results and Discussion

3.1 Insulating Layer Characterization

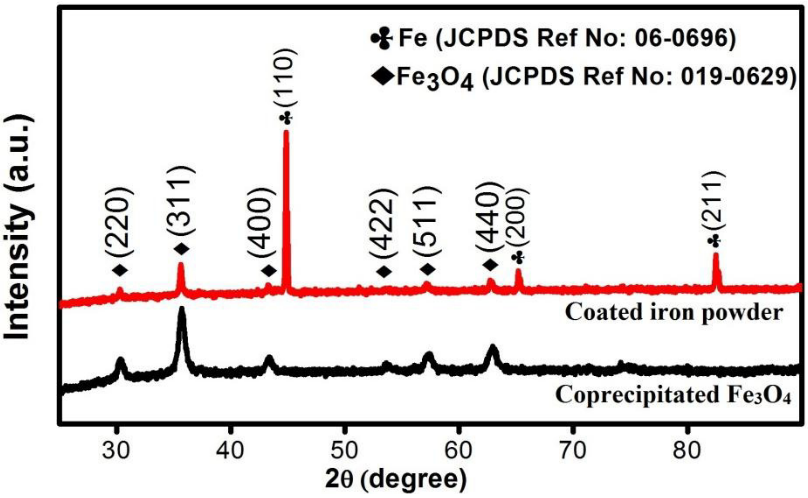

Figure 1: XRD patterns of the synthesized Fe_3O_4 and coated iron powder.

Figure 1 shows XRD patterns of coated iron powder composite and Fe_3O_4 powder samples. It can be seen that, diffraction peaks of the coated iron are consistent with the standard pattern for Fe_3O_4 (JCPDS Card No: 019-0629) and iron (JCPDS Card No: 06-0696) respectively indicating that iron powder was coated with Fe_3O_4. The XRD pattern of Fe_3O_4 showed very broad peaks, indicating the ultra-fine nature and nano crystallite size of the synthesized particles. Cubic single phase nano sized Fe_3O_4 powder had been obtained by the co-precipitation method.

The grain size of the Fe_3O_4 nanoparticles was calculated from the Scherrer formula:

$$D = \frac{K\lambda}{\beta \cos\theta} \tag{2}$$

Where β is the full width at half maximum (FWHM), D is the grain size in nm, θ is the corresponding Bragg angle , λ is the Cu-K_α X-ray wavelength (0.154 nm), and K is the shape parameter which is taken as 0.89 for Fe_3O_4. The grain size, calculated using the highest intensity peak (311) at $2\theta = 35.4°$, was 26 nm.

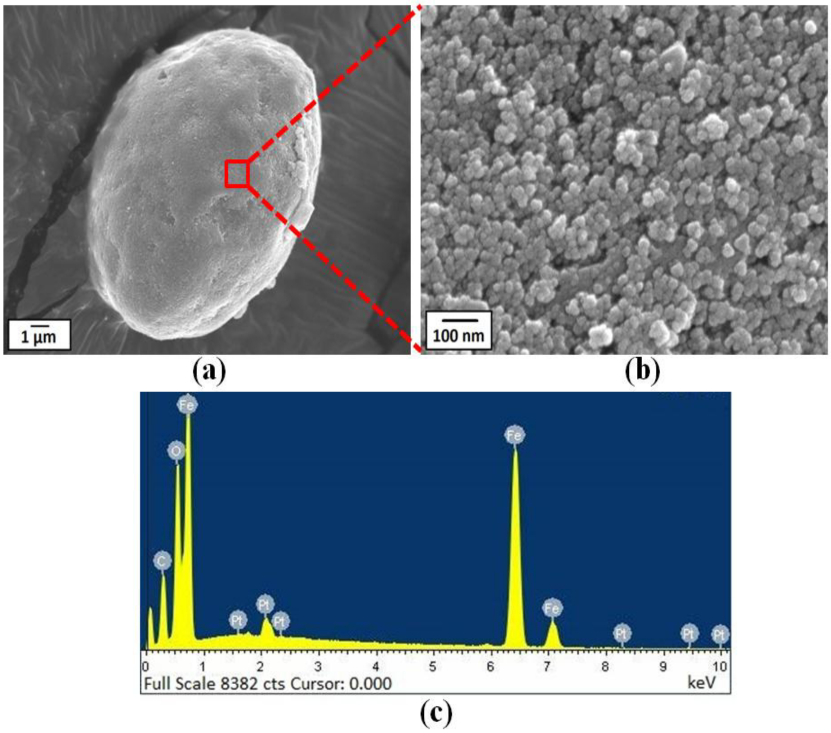

Figure 2: SEM images of (a) coated iron powder (b) coated iron powder surface (c) EDS analysis of coated iron powder.

Figure 2 shows the SEM images of coated iron powder and the performed energy dispersed analysis. We can see that iron powder is coated with nano sized Fe_3O_4 particles and that the coating is uniform. Making coating of nano Fe_3O_4 particles directly on iron powder is quite difficult. The epoxy resin binds the Fe_3O_4 nano particles to the iron surface and thus coats the iron powder. The epoxy resin also leads to the agglomeration of nano Fe_3O_4 particles which is observed in the SEM images. Figure 2(c) shows the EDS analysis of the Fe_3O_4 coated iron powder. The surface layer essentially consists of iron, oxygen and carbon. EDS also shows the presence of platinum because of metallic coating of the powder done before the SEM and EDS operation to reduce charging effects. Therefore from the XRD patterns analysis, SEM images and EDS analysis it is seen that the iron powder is coated uniformly with Fe_3O_4 nano particles.

3.2 DC Resistivity Measurement

Resistivity measurement of SMC is important to evaluate the insulation layer coated on iron powder. Four probe method is preferable for measuring resistivity since iron is a good conductor. In general resistivity measurement is done on rectangular bar or cylindrical sample of known dimension with uniform cross-section area. Unlike rectangular bar or cylindrical sample toroid shape is a closed geometry with uniform cross-sectional area. For this the current probes are connected at any two opposite points of the toroid that divide the toroid into two half portions (Figure 3(a)). The inner lengths of the two half portions of the toroid (L1 & L2) are equal.

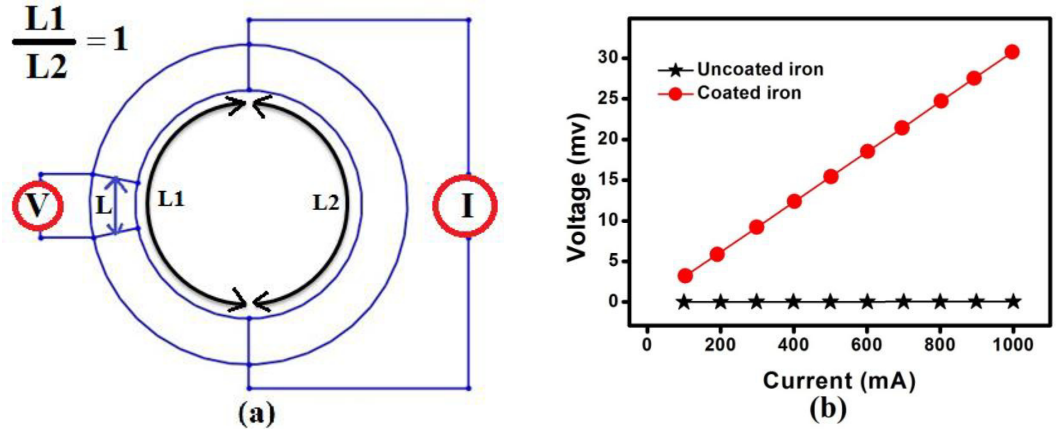

Figure 3: (a) Schematic diagram of toroid sample for resistivity measurement (b). I-V Characteristics of coated and uncoated iron.

Resistivity of toroids was measured since magnetic loss measurements are done on toroid shape samples. Equation employed for measurement of resistivity of toroid sample is given below [10].

$$\rho = \frac{2V}{I} \times \frac{a}{L} = \frac{R \times a}{L} \tag{3}$$

Where, ρ is Resistivity (Ω m), V is Voltage developed across voltage probe at a distance of L cm (v), I is Current supplied through current probes (A), a is cross sectional area (cm^2), R is the resistance of the toroid portion of length L and cross section a. I/2 is the current passing through half portion of the toroid and it is passing through a uniform cross section area, a, of the toroid.

A Keithley nanovoltmeter was used for the measuring the voltage drop, V, between the voltage sensing probes placed at a fixed distance, L, apart. A DC current of range of 100 to 1000 mA was passed and I-V characteristics were plotted keeping the voltage probes distance at 0.5 cm. Figure 3 shows the schematic diagram of toroid sample for resistivity measurement and I-V characteristics of coated and uncoated iron. The slope of the I-V graph gives the resistance R and equation (3) is used to calculate resistivity. From the figure 3(b) the resistivity of coated iron is 280 μΩm, where as for uncoated iron it is 0.2 μΩm.

3.3 DC & AC Magnetic properties

DC and AC hysteresis loops were recorded and core losses were measured on both uncoated and coated iron powder toroids using B-H Loop tracer. Figure 4 and Figure 5 shows the DC and AC hysteresis loops of uncoated iron powder and coated iron powder respectively. For uncoated iron the hysteresis loops were distorted as the frequency increased, due to the eddy currents induced by AC magnetic fields that opposes the applied field [11].

However for the coated iron due to its insulation on the surface the eddy current paths are limited to within the iron particle itself. Thus eddy currents are minimal in the coated iron powder. Thus results in undistorted and nearly superposed hysteresis loops as a function of AC frequency. This can be seen in the Figure 5. From figure 5 the superposition of the loops suggests that, the contribution of eddy current losses are small and that there is a good electrical insulation amongst the particles.

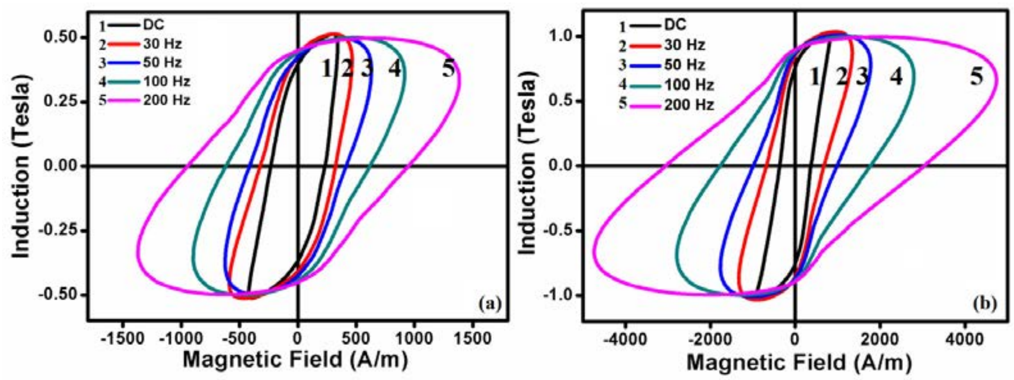

Figure 4: DC & AC hysteresis loops of uncoated iron powder at the induction level of (a) 0.5 Tesla (b) 1 Tesla (colour online).

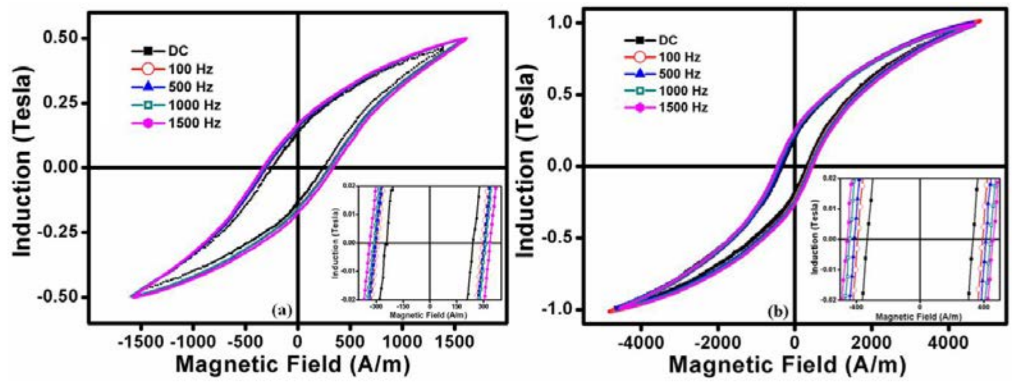

Figure 5: DC & AC hysteresis loops of coated iron powder at the induction level of (a) 0.5 Tesla (b) 1 Tesla (colour online).

The relation between core loss and frequency is shown in figure 6 at various frequencies and induction level of 0.5 Tesla and 1Tesla. For uncoated iron large change in core losses is seen due to dominant eddy current losses at higher frequency. The core losses are seen to be non-linear with the frequency. In the case of the coated iron due to its high resistivity, this relation is nearly linear, as eddy current losses are much smaller in magnitude. Coated and uncoated iron annealed at 600˚C exhibited a core loss of 91 W/kg and 693 W/kg respectively at 500 Hz and induction level of 1 Tesla.

The coated iron powder composite properties have been compared with the properties of a commercially available sample Somaloy 130i-1P, a product of Hoganas, Sweden [12]. Table 1 shows the comparison of properties between coated iron, uncoated iron and Somaloy130i-1P. Uncoated iron annealed at 600ºC has high maximum permeability (μ_{max}) and high magnetic induction due to the high density obtained. The coated iron on the other hand has lesser maximum permeability and lesser magnetic induction because of the insulation and pores present in the material. DC Induction value of coated iron powder and Somaloy130i-1P at 10000 A/m are 1.35 Tesla and 1.4 Tesla respectively, with

Core loss of 206 W/kg and 147 W/kg respectively at 1000 Hz and 1 Tesla. Utilization of the magnetic nature of Fe_3O_4 in the insulation yielded properties similar to Somaloy130i-1P. Epoxy resin was used in the composite to bind the nano Fe_3O_4 particles on to the iron powder.

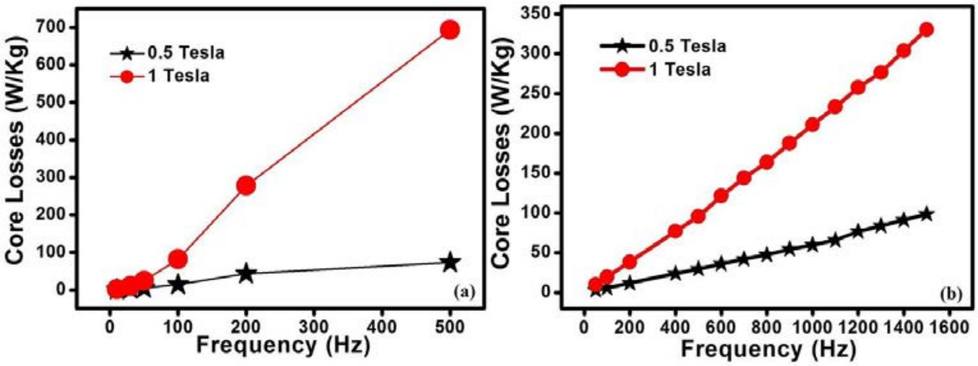

Figure 6: Core losses of (a) uncoated iron and (b) coated iron powder at the induction level of 0.5 Tesla and 1 Tesla.

Table 1: Comparison of properties between coated iron, uncoated iron and Somaloy130i-1P.

Sample	Density (g/cc)	Resistivity [μΩm]	Induction @ 10000 A/m [Tesla]	μ_{max}	Core Losses @ 1T [W/kg]		
					100 Hz	500 Hz	1000 Hz
Uncoated iron	7.6	0.2	1.60	1402	82	693	-
Coated iron	7.4	280	1.35	309	19	95	206
Somaloy130i-1P	-	8000	1.40	290	12	54	145

4 Conclusion

A new method to prepare coated iron powder has been attempted. SEM images showed the presence of uniform coating on iron powder. AC hysteresis loop studies of coated and uncoated iron powder showed the effect of the insulation coating. The core losses, induction and permeability of resulting coated iron were comparable with that of Somaloy130i-1P.

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