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High frequency properties of composite membrane with in-plane aligned Sendust flake prepared by infiltration method

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ABSTRACT

Composite membranes (with thickness around 100–200 μm) containing highly in-plane aligned Sendust flakes embedded in polyvinyl alcohol matrix were prepared with a novel infiltration method. As compared with tape-casting method, infiltration method results in enhanced magnetic permeability, which could be caused by better alignment and less porosity. Annealing process could modify the grain size, improve saturation magnetization and coercivity of Sendust flakes. Hence, the radio and quasi-microwave frequency permeability (between 10 MHz to 3 GHz) of composites membranes with annealed Sendust flakes could be enhanced significantly as compared with that of the as-prepared flakes. Infiltration method is especially suitable for composites with high concentration of flaky fillers. The composite membranes prepared have potential applications, including electromagnetic shielding, noise reduction and wave absorption.

1. Introduction

Sendust powder, a brittle magnetic alloy of iron, silicon and aluminum, was first produced in 1936 [1]. To enhance magnetic permeability for low frequency and low loss applications, Sendust flakes were prepared by warm rolling of sendust powder in 1957 [2]. Radio frequency and quasi-microwave magnetic permeability and its frequency dependency were investigated since 1999 [3–6]. It was proved that mechanical attrition is able to increase high frequency permeability of as-atomized Sendust flake [3]. The peculiar dual dispersion of permeability could be explained by the stress caused by attrition and increased magnetoelastic effect due to excess Fe by oxidation of Si and Al in the surface layer. Hence, better permeability characteristics could be obtained from a formula with less Fe content. The dispersion appeared in the lower frequency range (DII, around 100 MHz) could be correlated to a shape anisotropy of the flakes. In addition, the dispersion appeared in the higher frequency range (DIII, around 2 GHz) may be caused by the composition gradient structure consisting of a Fe-rich mother phase and a Si/Al-rich surface layer [4]. Noise suppressing effect (loss power over input power) up to 80% of a composite sheet made of forged Sendust flakes were measured with a microstrip line setup [4,5]. DC magnetic field could improve the electromagnetic-noise suppressing features through modifying magnetic resonance frequency [6].

Beside EM noise suppressing, Sendust flakes could also be used for EM wave absorption applications [7,8]. Co- or Ti-doped Sendust alloy was prepared with high-energy planetary ball milling method from melt-spun ribbons to improve the magnetic permeability and loss at GHz frequency, which are the key parameters in designing of high loss materials [7,9]. It was found that Ti doping increase the electrical resistivity and reduce the eddy current loss of magnetic composites in high frequency. The working frequency can be enhanced from 1.99 GHz to 2.45 GHz by substituting 1.5 wt% Ti for Fe [9]. Milling time and annealing temperature were also found to play important role in the nanocrystalline structure and magnetic properties of Co-doped Sendust alloy [7]. Attenuation loss (~10 dB) between 0.5 to 4 GHz of Sendust flakes composite was calculated from permeability and permittivity measured from toroidal samples [8].

Sendust flake composite were normally prepared using tape-casting method [3–6,10] or compacting molding method [8,9] from slurry of Sendust powder, polymer and solvent. The main purpose of the tape-casting processing is to align Sendust flakes of large aspect ratio within polymer matrix. The viscosity of Sendust flakes/acryl suspension was reported to be inversely proportional to the magnetic permeability of the resulting composites [10]. An anionic surfactant is effective way to lower the viscosity in certain concentration range, which results in better effective permeability. The reason could be the easier alignment of the flaky particles to the plain direction of sample sheet [10]. However, this method is not suitable for very high concentration. Infiltration method was recently proposed to prepare carbon nanotube membrane of high concentration of inclusions [11]. It

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solves the processing bottleneck (poor disperation and alignment) by separating the fabrication into two steps, namely, to prepare Sendust flakes paper by filtering them out from powder suspension in step one and to infiltrate polymer matrix into Sendust paper in step two.

Since thin paper-like sturctures containing Sendust flakes of good in-plane alignment could be made using the infiltration method, the main purpose of this paper is to fabricate Sendust flakes membrane with the infiltration method that has not been reported in literatures. To optimize magnetic permeability, the other purpose of this paper is to investigate the effect of annealing process on static and high frequency magnetic. Even though the effect of anealing time and temperature on grain size of Sendust flakes was already reported in [4,9], the anealing effect on high frequency magnetic properties is still not clear to the author's best knowledge.

2. Preparation and characterization

Sendust flakes prepared with attrition milling (with 9.59%Si, 5.88%Al and balance Fe) have mean diameter of 37 μm (measured with HORIBA, LA series) and thickness about 1 to 3 μm, as shown in SEM (JOEL JSM_6701F) image in Fig. 1. The as-prepared flakes were annealed in vacuum furnace at different treatment temperatures (from 300 °C to 700 °C) for 2 hours.

XRD (Rigaku Ultima IV) was used to analyze crystalline structure of annealed and as-prepared Sendust flakes. Slow scan rate (1 °/min) could increase the resolution of minor peaks. It was found the grain size increase significantly with treating temperature which agrees with the results reported in [4,9]. VSM (ADE Magnetics EV-9) were employed to measure hysteresis loops of annealed and as-prepared Sendust flakes with external field up to 2.1 T. Coercivity (H_c) and saturation magnetization (M_s) were obtained from hysteresis loops measured.

After annealing, 0.4 g flakes were dispersed in DI water using ultrasonication processor (Sonic VCX500) for 20 min. The ultrasonication process could break the particle agglomerations and disperse them to form water suspension. Right after that, the flakes were filtered out from the suspension with membrane filter (ISOPORE, size of pore: 0.4 μm). Then 1.5 ml PVA (polyvinyl alcohol) water solution (5 wt%) was dispensed uniformly on top of the thin flakes paper using syringe. The PVA solution will infiltrate into the porous sendust paper due to the negative pressure generated by a vacuum pump. The composite membrane of Sendust flakes and PVA solution was dried out in vacuum oven to remove water before peeling it off from the filter paper. The thickness of membrane prepared is about 100–200 μm and the diameter is 47 mm. The weight concentration measured by decomposing PVA inside membrane in a tube furnace at 300 °C for 2 hours is around 85 wt%. Fig. 1(a, b) show top and intersecion image of the Sendust membrane prepared using infiltration method. It can be seen that flakes regularly align in plane due to the strong pressing force from the vacuum pump, and the large aspect ratio (10∼20). It is not difficult to prepare membrane with larger thickness and size with longer processing time and larger fabrication facility. In fact, infiltration method is quite common for industry to prepare fiber-reinforced composites.

For comparison purpose, Sendust membrane with similar thickness was also prepared by tapecasting method. A bar-shape applicator was used to disperse the PVA and flakes slurry, by which membranes prepared have better surface smoothness and uniformity than that prepared manually. Fig. 1(c, d) show top and intersection view of the membrane prepared. It can be clearly seen from Fig. 1 that better in-plane alignment, smoother surface and denser pack could be realized with infiltration method.

High frequency permeability of the membranes prepared was measured with impedance analyzer (Agilent 4991 A and fixture 16454 A) from 0.01 to 3 GHz. The toroidal shape sample was cut from Sendust membrane with a specially designed metal cutter.

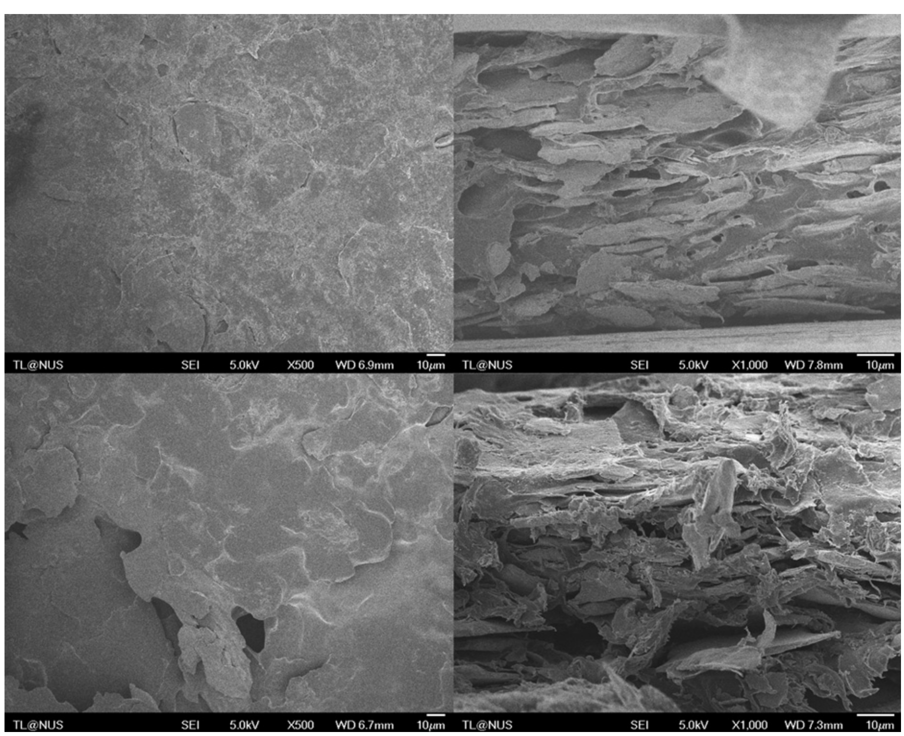

Fig. 1. SEM image of Sendust flakes composite prepared by bucky paper infiltration method, (a) top image and (b) intersection image and that prepared by tape casting method, (c) top and (d) intersection image.