Detonation Consolidation of NiFe/SiO$_2$ and Co/SiO$_2$ nanocomposites

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ABSTRACT

Consolidation of nanostructure magnetic particles is required not only for manufacturing bulk component, it is actually a fundamental requirement for obtaining novel magnetic properties from the material. Consolidation (assembly) of nanoparticles to full density without deteriorating their nanostructure (size and morphology) is a big challenge. Here we present the consolidation experiments of NiFe/SiO$_2$ and Co/SiO$_2$ nanocomposites via detonation consolidation. This approach is based on the explosive pressure created when an acetylene and oxygen mixture gas fires in a sample containing tube, the very high hypersonic propulsion force makes nanoparticles deposit onto the target. Depending on the powder morphology and operation conditions, the density of the consolidated sample can reach over 91% of the theoretical density of the bulk materials. X-ray diffraction experiments on the samples before and after consolidation indicate that the denotation consolidations can be optimized such that it does not cause any phase transition. However, a particle size increase was observed. Static magnetic studies carried out on the samples before and after detonation operation shows that the saturation magnetization does not. This indicates that the operation does not cause an oxidation of the nanopowders. These experiments show that detonation approach is a good candidate for consolidating magnetic nanoparticles.

INTRODUCTION

Nanocomposite fabrication provides a new opportunity to develop novel soft magnetic materials. In a metal/insulator nanocomposite system, its resistivity can be dramatically increased compared with conventional metallic alloys, leading to significantly reduced eddy current loss. Meanwhile, the exchange coupling between neighboring magnetic nanoparticles can overcome the anisotropy and demagnetizing effects, resulting in much better soft magnetic properties than conventional materials.$^{1,2}$ In pursuit of this idea, a large number of metal/insulator nanocomposite powder systems have been explored based on powder processing, which is advantageous for mass production of nanocomposite materials.$^{3-5}$ However, densifying nanoparticles into a bulk material while retaining their nanostructure is a big challenge. Among the consolidation approaches,$^{6,7}$ detonation compaction is a promising one.$^{8-12}$ The detonation gun spray process, referred to as the D-gun process, has been widely used in various industrial applications where a layer with high strength, high hardness and high density is required. In the D-gun process, the combustion of oxygen and acetylene is ignited using a sparking device and the ejected powder feedstock is semi-melted by the combustion heat. The droplets driven by the detonation wave are accelerated in the gun barrel toward the substrate at a high speed of about
800 to 1000 m/s. Thus a resultant layer can be formed with high bond strength and very low porosity due to the high kinetic energy and high velocity. In this work, a consolidation study of NiFe/SiO$_2$ and Co/SiO$_2$ using detonation compaction is presented.

EXPERIMENTAL DETAILS

(Ni$_{75}$Fe$_{25}$)$_v$/(SiO$_2$)$_{1-v}$ and Co$_v$/(SiO$_2$)$_{1-v}$ nanocomposites, where $v$ denotes the volume fraction of metal phase in the composites, were synthesized using a wet chemical solution technique. Solutions of Ni-, Fe- and Si-containing precursors were prepared separately in alcohol with controlled pH, reaction time, and temperature, to form precomposite precipitates in solution. The precipitated nanoparticle suspensions were dried in an oven to obtain a precomposite powder. The precomposite powder was then thermochemically converted into a metal-ceramic (NiFe/SiO$_2$ or Co/SiO$_2$) nanocomposite powder in the presence of a reducing agent at temperatures from 400 to 700 °C. The nanocomposites were passivated to prevent oxidation.

The consolidation of NiFe/SiO$_2$ and Co/SiO$_2$ was performed using a custom-built D-gun facility. This facility consists an aluminum disk (10-15 mm in diameter) with a central post (6 mm in diameter) which was used as substrate. The substrate was degreased in acetone and surface coarsened by sand blasting prior to D-gun spraying. The substrate was inserted into a cylinder holder (20 mm I.D.) that was fixed on a stand. In the D-gun operation process, nano-NiFe/SiO$_2$ or Co/SiO$_2$ powder was fed into the barrel. Then the barrel was filled with a O$_2$ and C$_2$H$_2$ gas mixture, and the mixture was ignited by means of a spark plug. The ignition caused combustion and detonation of the gas mixture, which forced the nanopowder onto the Al substrate to form a deposit. In this process, the main variables included O$_2$ and C$_2$H$_2$ flow rates and the distance between the powder feedstock and substrate.

The crystal structure was determined by x-ray diffraction (XRD) experiments. Also, the particle sizes of the as-synthesized nanoparticles and the grain sizes of the consolidated samples were calculated from the linewidth of the main XRD line using the Scherrer equation. The morphology of the consolidated samples was checked by scanning electron spectroscopy. The densification quality was characterized by a packing density, which is defined as the ratio of the measured density, $d$, of the densified sample to its theoretical density, $d_{TD}$. The value of $d$ was measured using Archimede’s law while the theoretical density of (NiFe)$_v$/(SiO$_2$)$_{1-v}$ or Co$_v$/(SiO$_2$)$_{1-v}$ was calculated according to the volume fractions of the metal and SiO$_2$ phases. The static magnetic properties were studied using a Quantum Design SQUID magnetometer. Initial complex permeability, $\mu = \mu' - j\mu''$, vs frequency curves were measured using an impedance meter on the as-compacted samples and annealed samples.

RESULTS AND DISCUSSION

Figure 1 shows XRD patterns for Ni$_{75}$Fe$_{25}$)$_{0.5}$/(SiO$_2$)$_{0.5}$ (a) and Co$_{0.5}$/(SiO$_2$)$_{0.5}$ obtained on the as-synthesized nanoparticle and on the D-gun consolidated sample. As shown in the figure, the XRD peak 2θ values for the samples before and after consolidation are the same, indicating that the D-gun operation does not cause any change in the structure. However, a significant increase in the grain size was observed after consolidation as seen by the sharper peaks. For Ni$_{75}$Fe$_{25}$)$_{0.5}$/(SiO$_2$)$_{0.5}$, the average particle size changed from 16 nm to 44 nm during
consolidation. For Co0.5/(SiO2)0.5 system, a increase of grain size from 30 to about 50 nm was resulted as shown in Fig. 1b.

Figure 2 shows a scanning electron microscopy (SEM) image of a D-gun consolidated (Ni75Fe25)0.5/(SiO2)0.5 sample. As shown in the figures (particularly in Fig. 2a), the D-gun consolidated samples possess a layered structure with a thickness of about 0.1 mm. Each layer corresponds one detonation. Within each layer, the powder is compacted densely, while the pores exist mostly in the interface region between neighboring layers. The thickness of the layer is determined by the mass of powder in each detonation. It was found that by increasing the mass of the powder for each detonation, more efficient compaction was achieved in that the thicker layer was attained. The highest packing density achieved is 90% for (NiFe)v/(SiO2)1-v, and 0.85% for Cov/(SiO2)1-v. It was found that the packing density is sensitively dependent on the flow rates of O2 and C2H2, the powder morphology and the spacing between the powder and the substrate. For instance, when nanopowders are agglomerated into large particles with homogeneous particle size distribution, more densely packed green component can be obtained.

Fig. 1. XRD patterns of (Ni75Fe25)0.5/(SiO2)0.5 (a) and Co0.5/(SiO2)0.5 (b) samples before and after D-gun consolidation.

Figure 2 shows a scanning electron microscopy (SEM) image of a D-gun consolidated (Ni75Fe25)0.5/(SiO2)0.5 sample. As shown in the figures (particularly in Fig. 2a), the D-gun consolidated samples possess a layered structure with a thickness of about 0.1 mm. Each layer corresponds one detonation. Within each layer, the powder is compacted densely, while the pores exist mostly in the interface region between neighboring layers. The thickness of the layer is determined by the mass of powder in each detonation. It was found that by increasing the mass of the powder for each detonation, more efficient compaction was achieved in that the thicker layer was attained. The highest packing density achieved is 90% for (NiFe)v/(SiO2)1-v, and 0.85% for Cov/(SiO2)1-v. It was found that the packing density is sensitively dependent on the flow rates of O2 and C2H2, the powder morphology and the spacing between the powder and the substrate. For instance, when nanopowders are agglomerated into large particles with homogeneous particle size distribution, more densely packed green component can be obtained.

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Fig. 2. SEM image of D-gun consolidated (Ni75Fe25)0.5/(SiO2)0.5 (a) and (Ni75Fe25)0.5/(SiO2)0.5 (b) samples
magnetization measurement as any oxidation will cause a reduction of saturation magnetization. Static magnetic studies carried out on the samples before and after the D-gun process resulted in essentially the same saturation magnetization values. This indicates that the D-gun operation does not cause an oxidation of the nanopowders.

Figure 3 shows the initial complex permeability of the D-gun consolidated (Ni$_{75}$Fe$_{25}$)$_{0.5}$/(SiO$_2$)$_{0.5}$ and Co$_{0.5}$/(SiO$_2$)$_{0.5}$ samples as a function of frequency. For bulk Ni$_{75}$Fe$_{25}$ alloy, its initial permeability should be of $10^3$; while for fcc Co it is of $10^3$. In comparison with these values, the consolidated (Ni$_{75}$Fe$_{25}$)$_{0.5}$/(SiO$_2$)$_{0.5}$ and Co$_{0.5}$/(SiO$_2$)$_{0.5}$ samples possess much smaller permeabilities. The packing densities for these two samples are 90% and 77%, respectively. In this case the demagnetizing factor may have larger influence on the magnetic softness than the exchange coupling. This may be the major reason that both samples possess relatively low permeabilities. On the other hand, both samples show a very good frequency response. If the SiO$_2$ coating were destroyed during consolidation, direct metal-metal contact would take place, the eddy current would cause $\mu'$ to decrease with frequency much faster than that shown in Fig. 3, and $\mu''$ would reach a maximum at a lower frequency, roughly in the kHz range for a 0.1 mm thick layer. As shown in Fig. 3, however, $\mu'$ is essentially flat and $\mu''$ is still small up to 2 MHz for (Ni$_{75}$Fe$_{25}$)$_{0.5}$/(SiO$_2$)$_{0.5}$ and 13 MHz for Co$_{0.5}$/(SiO$_2$)$_{0.5}$. This indicates a good insulation of the metal particles by SiO$_2$.

![Graphs showing permeability as a function of frequency for D-gun consolidated samples](a) and Co$_{0.5}$/SiO$_2$ samples (b).

**CONCLUSION**

This study shows that using the detonation consolidation approach, metal/insulator nanocomposite particles can be consolidated into a bulk component with very high packing density while the nanostructure of the particle and magnetic moment can be essentially retained. It does not cause phase transition and oxidation. Therefore, detonation compaction is a promising approach for consolidating magnetic nanoparticles.
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REFERENCES