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Investigation of oxidation resistance of magnetic power coated with silicone

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Abstract

In this work the oxidation resistance of a magnetic material of the neodymium–iron–boron (NdFeB)-type coated with silicone has been investigated by thermogravimetric analysis (TGA) and differential thermal analysis (DTA) techniques. A special spin-coating technique has been used to coat the surface of the magnetic powder NdFeB. The results of TGA and DTA indicate that the oxidation resistance of the surface-treated NdFeB powder is markedly improved, while its magnetic properties do not change much compared to those of untreated material.

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Keywords: Magnetic powder; NdFeB; Silicone; Spin coating; Oxidation resistance

It is well known that the magnetic material of the neodymium–iron–boron (NdFeB) type has extremely large magnetic force [1]. This magnetic material can be obtained at low price because it contains a large amount of iron. However, this type of magnetic material is readily oxidized. Because its major component is iron, the magnetic material will corrode and rust in the presence of water. As a result, not only the magnetic force of a resin-bonded permanent magnet will be greatly decreased, but the rust inside the magnet will eventually destroy the magnet itself.

In order to solve these problems, many attempts have been made to protect the magnetic powder from oxidation [2–4]. However, no satisfactory anti-rusting properties have been provided. Recently, it is reported that a surface-treated magnetic powder with excellent oxidation resistance and excellent moisture resistance is obtained by treating a rare-earth iron magnetic powder with a treatment agent containing alkali-modified silica particles as a major component [5].

The purpose of this work is to study the oxidation resistance of the magnetic powder NdFeB surface-treated with spin-on silicone by means of thermogravimetric analysis (TGA) and differential thermal analysis (DTA) methods.

1. Experimental

The NdFeB magnetic powder used in this work was supplied by Jilin magnetic material plant

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(China) with an average particle size of 5 μm. The ethanol solution of silicon (18 vol%), as a surface treatment agent for NdFeB magnetic powder, was prepared by continuous reactions of hydrolysis and polycondensation of tetraethoxysilicone (TEOS) in ethanol solvent with the hydrochloric acid as catalyst

$$(C_2H_5O)_4Si + H_2O = (C_2H_5O)_3SiOH + C_2H_5OH\,,$$

$$2(C_2H_5O)_3SiOH = (C_2H_5O)_6Si_2O + H_2O\,.$$

By controlling the ratio of O/Si in ethoxy silicone near 1 or smaller than 1, an ethanol solution of silicone polymer with linear structure was obtained, which was used as the surface-treatment agent for the NdFeB magnetic powder.

A special spin-coating technique was used to treat the surface of the NdFeB magnetic powder. Unoxidized NdFeB magnetic powder was added to the ethanol solution of silicone prepared as above. In order to disperse the powder uniformly in the solution, ultrasonic wave stirring was used. Subsequently, the mixture of powder and the silicone solution was moved into a special filter in which the residual solution was spun off at high spinning speed, depositing a thin layer of silicone on the surface of the powder. The thickness of the coating layer was controlled by the spinning speed of the filter. The coating layer thickness of 0.5 μm was obtained at a spinning speed of 2500 rpm. The surface-treated NdFeB magnetic powder was then dried in a vacuum oven at 60°C for 30 min followed by 110°C for 1 h.

2. Results and discussion

Fig. 1 shows the infrared spectra of the silicone coated on silicon substrates heat-treated at 60 and 110°C, respectively. The absorption bands at 1096.1, 1194.1 and 3353.1 cm^{-1} represent Si–O, Si–O–Si and –OH stretching vibrations, respectively [6]. When the heat-treatment temperature increases, the absorption intensities of the bands at 1096.1 and 1194.1 cm^{-1} increase, while the intensity of the band at 3353.1 cm^{-1} decreases. This implies that, during the heat treatment, further dehydration occurs forming Si–O and Si–O–Si groups, and, thus, resulting in increased oxidation resistance of the silicone.

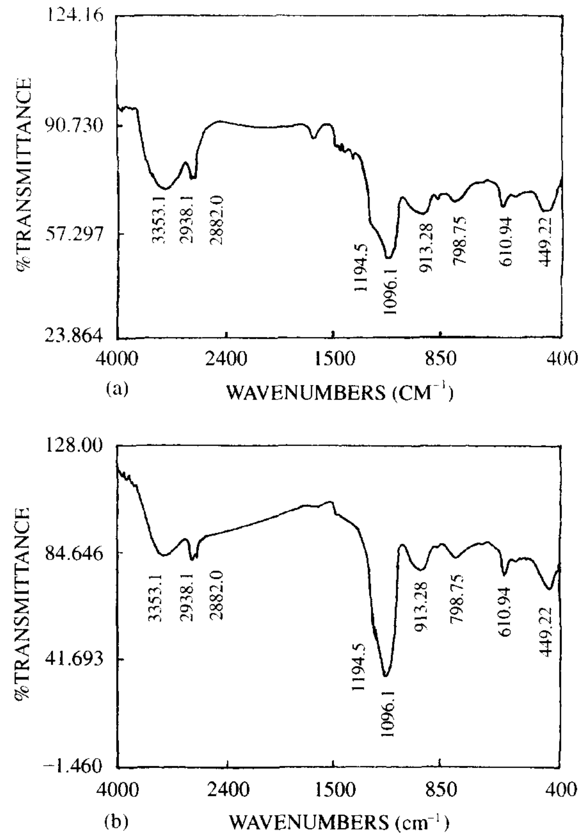

Fig. 1. Infrared absorption spectra of silicone coated on silicon substrates: (a) heat-treated at 60°C and (b) heat-treated at 110°C.

Fig. 2 shows the results of thermogravimetric analysis (TGA) of uncoated and coated NdFeB magnetic powders. During the heating process, weight decrease takes place at temperatures below 150°C due to additional dehydration of the magnetic powder. It is obvious that the moisture retained by the uncoated magnetic powder is much higher than that of coated powder. It should be pointed out that, for uncoated magnetic powder, the weight increases markedly (Fig. 2b) when the temperature is above 300°C. Unambiguously, the

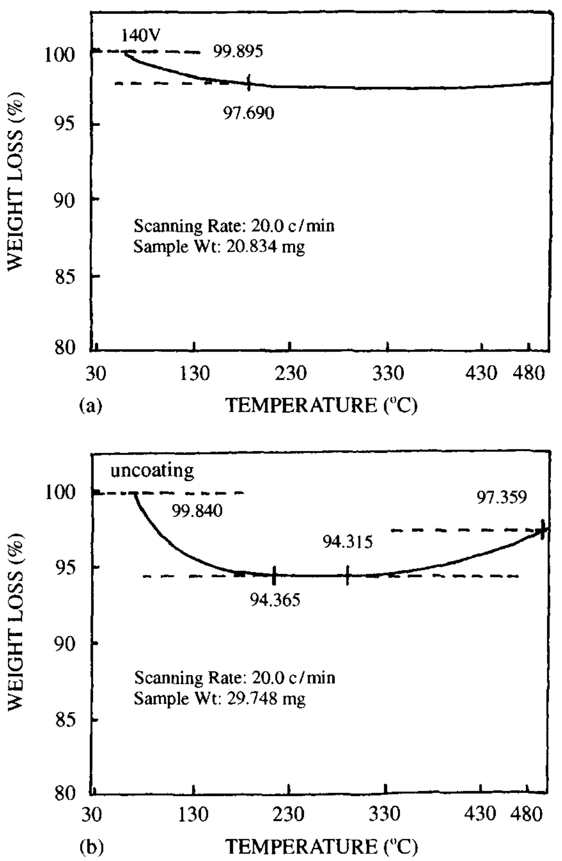

Fig. 2. Results of TGA on NdFeB magnetic powder coated (a) and uncoated (b) with silicone.

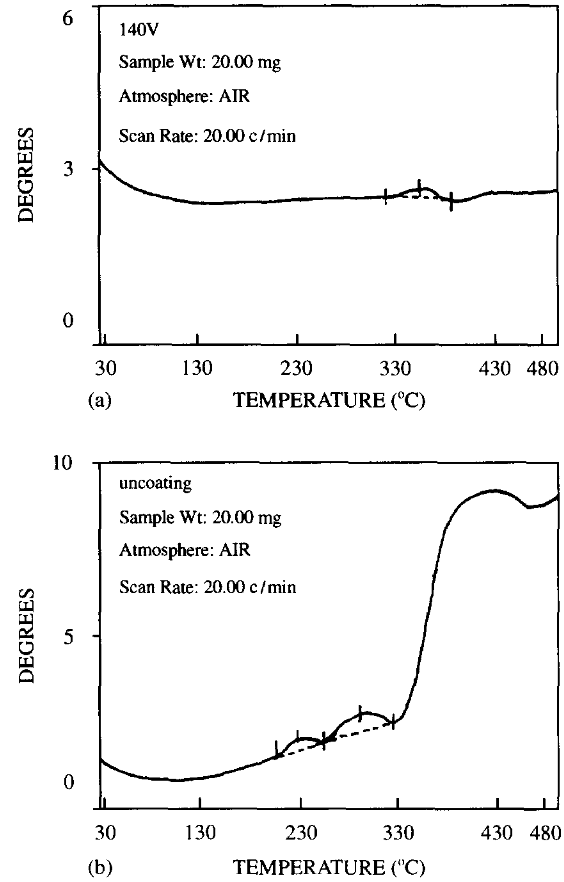

Fig. 3. Results of DTA of NdFeB magnetic powder, coated (a) and uncoated (b).

weight increase is the result of oxidation of the powder at higher temperatures. However, the weight of the coated NdFeB powder almost does not increase until the temperature is up to 480°C (Fig. 2a). This implies that the coated NdFeB magnetic powder exhibits excellent oxidation resistance at higher temperature.

The results of differential thermal analysis (DTA) of coated and uncoated NdFeB magnetic powder are shown in Fig. 3. For coated magnetic powder, only a small exothermic peak at the temperature of 350°C has been observed (Fig. 3a), which should be attributed to silicone solidification and conversion into inorganic SiO_2. However, the uncoated NdFeB magnetic powder exhibits three exothermic peaks at 225, 290 and 420°C, respectively (Fig. 3b). The peak at 225°C is caused by the oxidation of neodymium, while the peak at 420°C refers mainly to the oxidation of iron. As for the one at 290°C, impurity or phase transformation probably is the reason. It should be pointed out that the exothermic peak at 420°C for the uncoated powder is very large, which implies that the iron in the uncoated NdFeB powder is readily oxidized at higher temperature. The above DTA results further confirm that the coated NdFeB powder has excellent oxidation resistance.

Fig. 4 shows the results of SEM observation of the coated and uncoated NdFeB powder. The coated layer of silicone on the surface of NdFeB magnetic powder is uniform and densified (Fig. 4b). Also, the surface-treated powder exhibits fine

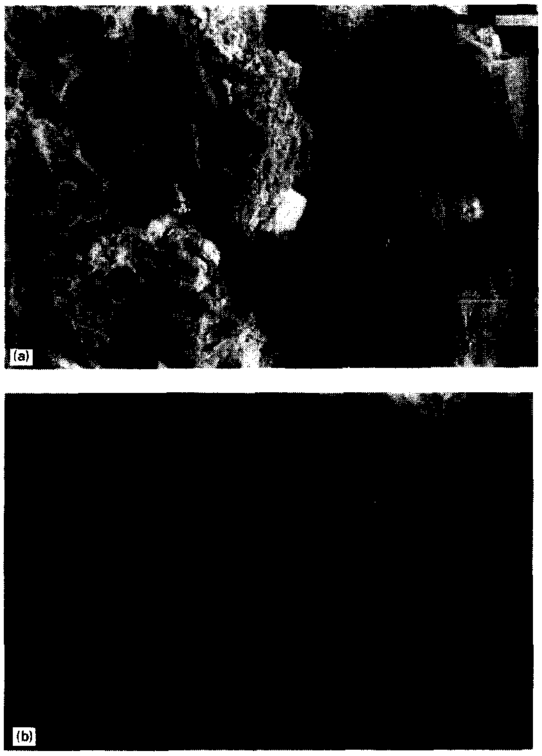

Fig. 4. SEM picture of uncoated (a) and coated (b) NdFeB magnetic powder.

dispersity compared with that of uncoated powder (Fig. 4a).

Finally, in order to compare the magnetic properties of the coated NdFeB magnetic powder with that of uncoated powder, resin-bonded cylindrical magnets with the size of ∅ 10 × 10 mm were prepared. The magnetic properties of the cylindrical magnets are shown in Table 1. It is obvious that little change of the magnetic properties of the coated powder is observed, compared to those of uncoated powder.

Table 1
The magnetic characteristics of coated and uncoated NdFeB magnet powder

	B_r (kG)	BH_c (kOe)	BH_{max} (mgOe)
Uncoated	5.4	4.1	5.6
Coated	5.2	4.0	5.5

3. Conclusions

(1) Silicone solution in ethanol prepared by TEOS hydrolysis and polycondensation, is an excellent surface treatment agent for NdFeB magnetic powder.

(2) The spin-coating technique is very useful for surface treatment of the magnetic powder, forming a uniformly thin layer (about 0.5 μm) of silicone on the surface of the NdFeB powder.

(3) The NdFeB magnetic powder coated with silicone has excellent oxidation resistance and moisture resistance.

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