

MAGNETIC PROPERTIES OF CARBON COATED IRON NANOCRYSTALLITES EMBEDDED IN CARBON MATRIX

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Introduction

Carbon materials are made via pyrolysis and carbonization of the organic precursor. By the addition of foreign materials to the organic precursor, structure and properties of the resultant carbon material are known to be greatly influenced [1-3]. Addition of transitional elements during the pyrolysis process results in formation of structures like nanotubes, carbon nanocapsules etc. to catalytic graphitization of the amorphous carbon, depending on the preparation method.

In this paper, structure and magnetic properties of as made carbon-coated iron nanoparticles are investigated. The compositions are prepared by chemical vapour deposition of ferrocene and maleic anhydride.

Experimental

The carbon-coated iron nanocrystallites embedded in carbon matrix are prepared by chemical vapour deposition of ferrocene and maleic anhydride. The samples are prepared at two different preparation temperatures mainly 900°C and 980°C. The iron content in the compositions is varied by changing the molecular weight percentage of ferrocene with respect to maleic anhydride. The compositions are prepared for 5%, 10%, 20% and 50% of ferrocene molecular weight for both the temperatures. A single zone wire wound furnace is used for the pyrolysis of organic precursors in a 50cm long, 10mm diameter pre-evacuated, and argon purged quartz tube.

The powdery samples obtained after 5hrs of pyrolysis process were collected and used for structural characterization and magnetic properties study. Powder X-ray diffraction on the composition was performed using CuK α radiation on a Philips X'pert Diffractometer. Magnetization curves were obtained in a vibrating sample magnetometer (VSM), at room temperature. SQUID magnetometer was used to plot the hysteresis loop for some of the samples.

Results and Discussion

The structural characterization of the samples by powder x-ray diffraction (fig.1) shows sharp increase in the graphite (002) peak with increase in pyrolysis temperature and

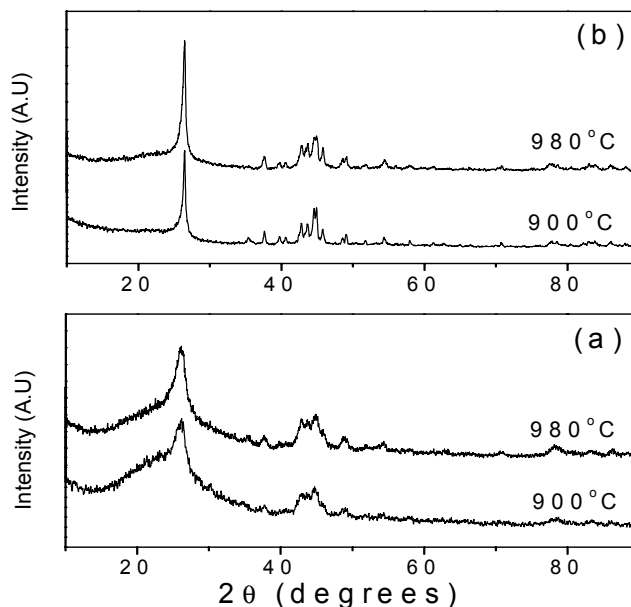


Figure 1. X-ray diffraction pattern for the powder composition of (a) 5% and (b) 50% ferrocene composition samples prepared at 900 °C & 980 °C

iron content in the precursor. The graphitization of the amorphous carbon matrix is occurs due to the catalytic activity of the transitional element, Fe. The process of formation of intermediate compound Fe_3C and its recrystallization as graphite and iron crystal is well documented in literature [4]. The FWHM for the (002) peak decreases with increase in ferrocene percentage in precursor and pyrolysis temperature. The crystallite sizes are calculated using the Debye Scherrer equation,

$$L_C = K\lambda / \beta \cos\theta \quad (1)$$

where K is the shape factor ($=0.9$), λ is the wavelength of X-ray (1.5406 \AA), β is the FWHM at the Bragg angle, θ . The graphite crystallite size (L_C) increases with ferrocene percentage and pyrolysis temperature and varies from 4nm to 30nm.

Fig. 2 shows the dependence of the saturation magnetization (M_S) and remanent magnetization (M_R) as the ferrocene percentage and pyrolysis temperature. M_R/M_S value of the powder samples is also shown. Value of saturation magnetization does not show a direct dependence on the ferrocene percentage. If comparison is made via preparation temperature 980°C samples shows higher M_S value for all the compositions except 50% ferrocene sample. This decrease in value is due to the effect of graphite on the magnetization behavior of the embedded nanoparticles as follows. Graphite is diamagnetic, with $-21.5 \times 10^{-6} \text{ emu/g}$ parallel to the magnetic field and $-0.5 \times 10^{-6} \text{ emu/g}$ perpendicular to the magnetic field [5]. With increasing amount of the graphite, this effect may become enhanced. After the preparation of the 50% ferrocene composition at 980°C, the effect of graphite apparently decreases the saturation magnetization. The trend of the M_S value along the X-axis of the graph also shows dependence on the

extent of graphitization of the carbon matrix with increase in iron percentage. The ferromagnetic behaviour exhibited by the present sample is due to the iron carbide (Fe_3C) nanocrystallites embedded in the carbon matrix. The Curie temperature (T_C) of Fe_3C crystal is around 220°C and has a saturation magnetization value of 140 emu/g .

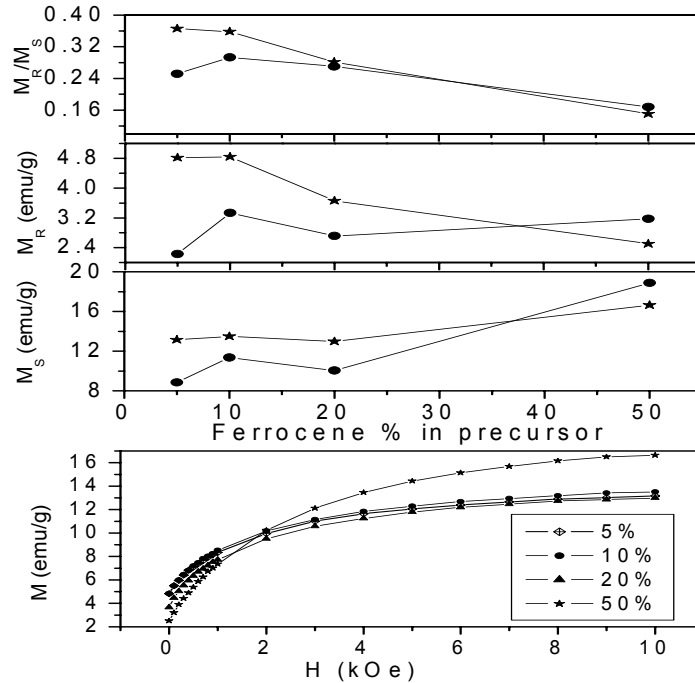


Figure 2. Plot of remanence ratio, M_R/M_S ; remanence magnetization, M_R ; saturation magnetization, M_S as a function of ferrocene content in the precursor and the demagnetization curve for the 980°C prepared samples

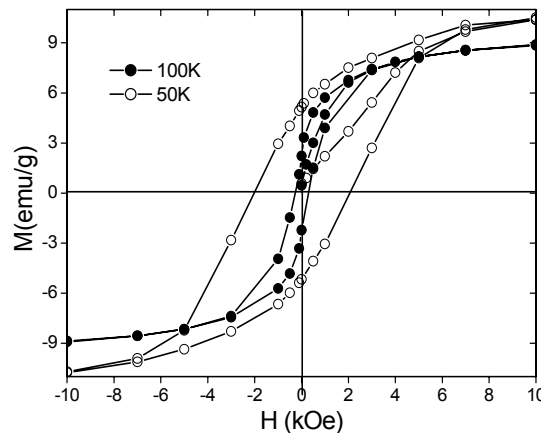


Figure 3. Hysteresis loop for the 5% ferrocene composition powder sample prepared at 900°C at temperatures 300K and 50K

The hysteresis loop (Figure 3) is plotted using SQUID magnetometer on two of the 900°C prepared sample. The coercivity values for 5% & 50% ferrocene composition at

room temperature are 270 and 388 Oe respectively. The value observed is consistent with the nucleation model of domain wall proposed by Kittel [6]. At low temperature the coercivity value of the two samples shows 1/d dependence according to the formulation by Chen et. al. [7] having values 2 kOe and 1.25 kOe respectively.

Conclusions

Iron nanoparticles embedded in carbon matrix are prepared from pyrolysis of maleic anhydride and ferrocene at high temperatures. The structure and magnetic properties of the sample depends on the preparation temperature and amount of ferrocene in the precursor. Graphitization of the amorphous carbon by the catalytic action of the transitional element Fe has a direct influence on the ferromagnetic behaviour of the composition. A high coercivity value ~2kOe is seen for the lowest iron concentration sample in the nanoparticle assembly.

References

- [1] Song H, Chen X. Large scale synthesis of carbon-encapsulated iron carbide nanoparticles by co-carbonization of durene with ferrocene. *Chem. Phys. Lett.* 2003;374:400-04
- [2] Sano N, Akazawa H, Kikuchi T, Kanki T. Separated synthesis of iron-included carbon nanocapsules and nanotubes by pyrolysis of ferrocene in pure hydrogen. *Carbon* 2003;41:2159-79
- [3] Liu X, Huang B, Coville NJ. The Fe(CO)₅ catalyzed pyrolysis of pentene: carbon nanotube and carbon nanoball formation. *Carbon* 2002;40:2791-99
- [4] Yudasaka M, Kikuchi R. Graphitization of Carbonaceous Materials by Ni, Co and Fe. Yoshimura S, Chang RPH, Editor. *Supercarbon*, Springer, 1998:99-105
- [5] Wang ZH, Zhang ZD, Choi CJ, Kim BK. Structure and magnetic properties of Fe(C) and Co(C) nanocapsules prepared by chemical vapor condensation. *J. Alloys Comp.* 2003;361;289-93
- [6] Kittel C. Physical theory of ferromagnetic domains. *Rev. Mod. Phys.* 1949;21:541-83
- [7] Chen C, Hitakami O, Okamoto S, Shimada Y. Surface anisotropy in giant magnetic coercivity effect of cubic granular FeCo/SiO₂ and NiCo/SiO₂ films: A comparison with Neel's Theory. *J. Appl. Phys.* 1999;86(4):2161-65