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Enhanced remanence behavior in mechanically alloyed $SmCo_5$

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Abstract

Sm–Co powders produced by a mechanical alloying process were investigated in terms of their structure, morphology and magnetic properties. Remanence enhancement, with M_r/M_s ratios up to 0.69 were observed and the high coercivity was found to be related to the development of the $SmCo_5$ phase.

Keywords: Mechanical alloying; Exchange-coupling; SmCo; Magnetic material

1. Introduction

Hard magnetic nanocrystalline materials consisting of a mixture of hard and soft phases have been the subject of considerable interest. These materials, whilst being microstructurally isotropic, exhibit an enhanced remanence as a result of magnetic exchange coupling between the hard and soft magnetic phases. The materials can be produced either by melt spinning or by mechanical alloying, combined with a suitable heat treatment.

Mechanical alloying was first developed as a means of manufacturing oxide dispersion strengthened super alloys [1], but has more recently found applications in the formation of amorphous alloys [2] and magnetic materials [3]. Although initially, mechanical alloying, like melt spinning, was used primarily as a method for fabricating conventional isotropic magnetic materials which could either be plastic bonded or used as a precursor for die upset forging, its potential for producing exchange coupled materials was not realised until later. The first evidence for exchange coupling was in NdFeB alloys reported by Clemente et al. [4] for a near stoichiometric melt spun $Nd_2Fe_{14}B$ and by Coehoorn et al. [5] for a melt spun NdFeB material which mixed the hard magnetic $Nd_2Fe_{14}B$ with soft magnetic Fe_3B and α-Fe. Subsequent studies by Ding et al. [6] using $Sm_2Fe_{17}N_x$ with an excess of α-Fe showed that it was possible to obtain exchange-coupled materials showing enhanced remanences by mechanical alloying. Materials with BH_{max} values of 20 MGOe were produced, similar energy products have been obtained by melt spining $Nd_2Fe_{14}B+\alpha$-Fe ribbons [7,8].

Although the literature contains some reports of mechanical alloying of SmCo-based alloys in order to develop isotropic [9] and exchange-coupled [10,11] magnets this material has not received the same attention as either the NdFeB or SmFeN materials.

In this paper we report on a study of mechanically alloyed materials with compositions close to $SmCo_5$.

2. Experimental

Mechanically alloyed samples were prepared from samarium (~130 μm) and cobalt (~70 μm) powders of 99.9% purity. Starting compositions in the range Sm_xCo_5, where $x=0.95$–1.25, were mechanically alloyed for 3000 min in an argon atmosphere of 1 bar using four 22 mm steel balls in an ASI Uni Ball Mill. Powders were subsequently heat treated in an argon atmosphere using a conventional vacuum furnace. Structures were determined by X-ray diffraction using a Philips PW1710 diffractometer and CuKα radiation. Magnetic measurements were carried out at room temperature using a Manics DSM.

3. Results and discussion

All samples were investigated after mechanical alloying in order to determine that the materials had become amorphous. X-ray diffraction patterns indicated only very broad peaks of low intensity similar to those observed by other workers [10]. The material exhibited a dark grey colour and the particle size and morphology were investigated using a JEOL 5800 scanning electron microscope

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(SEM). A relatively narrow particle size distribution with most particles falling within the range 10–30 μm was observed, there is little evidence for significant amounts of debris. A close up view of one particle (see Fig. 1) confirms that each particle is basically spherical in shape and appears to be made up of layers of material in agreement with previous observations for mechanically alloyed materials [12].

During the process of mechanical alloying a significant amount of alloying occurs not just between the particles themselves but also between the material and the mechanical alloying chamber. As a result of this it is often not possible to retrieve all of the initial charge. In the course of these experiments it was found that 2 or 3 g of material from the initial charge of 15 g could not be removed using any non abrasive technique. In order to make certain that no significant change in the chemical composition was taking place as a result of preferential alloying of one element to the walls of the chamber, chemical analysis was carried out on all of the mechanically alloyed samples. The graph, Fig. 2 shows slight preferential loss of samarium in the order of 1 wt. %.

Since the experimental work involved dealing with fine powders of high reactivity, oxygen analysis was carried out on the samples mechanically alloyed and heat treated at 875°C. The graph, Fig. 3 shows oxygen levels in the region of 0.4–0.6 wt. %, typical of that observed in other forms of processing rare earth-transition metal materials.

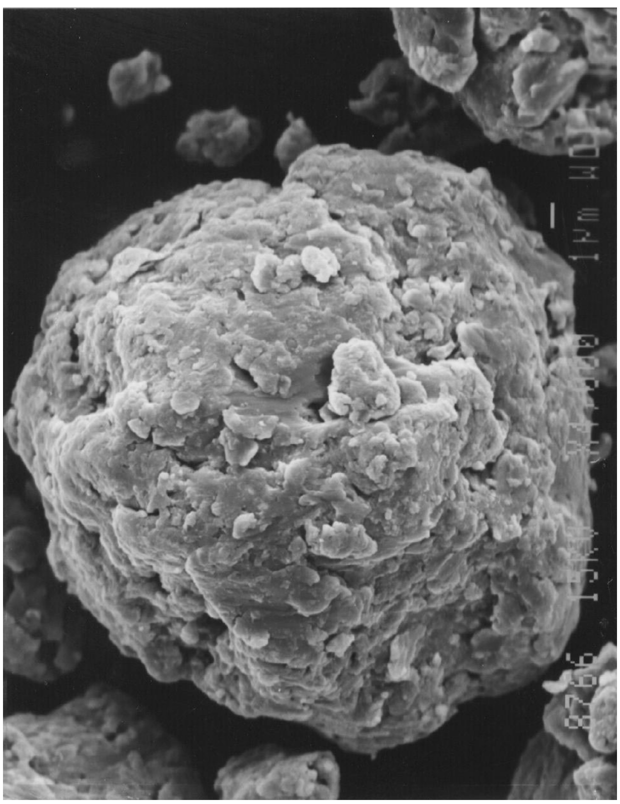

Fig. 1. Single particle of mechanically alloyed $Sm_{1.15}Co_5$.

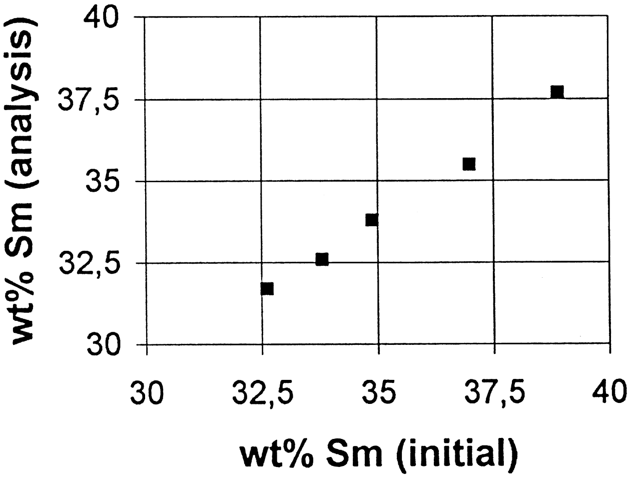

Fig. 2. Comparison of samarium contents of samples before and after the mechanical alloying process.

The slight increase in the degree of pick up with higher rare earth containing materials is consistent with the formation of Sm based oxides. Measurements relating to the amount of iron picked up during the experiment were not undertaken, experience from earlier studies on similar materials has shown that significant quantities are not observed.

The mechanically alloyed samples in the range Sm_xCo_5 where x=0.95–1.25 were each heat treated for 2 h at the following temperatures, 575°C, 675°C, 775°C and 875°C, the heating and cooling rates were 5°C/min and the furnace tube was filled with argon at a pressure of 1 bar after pre-evacuation.

X-ray diffraction was carried out using CuKα radiation. In Fig. 4 the traces from the as-mechanically alloyed sample of $Sm_{1.15}Co_5$ as well as $Sm_{1.15}Co_5$ samples heat treated at 575°C, 675°C, 775°C and 875°C are shown. The un-heat treated sample shows a very flat trace with broad

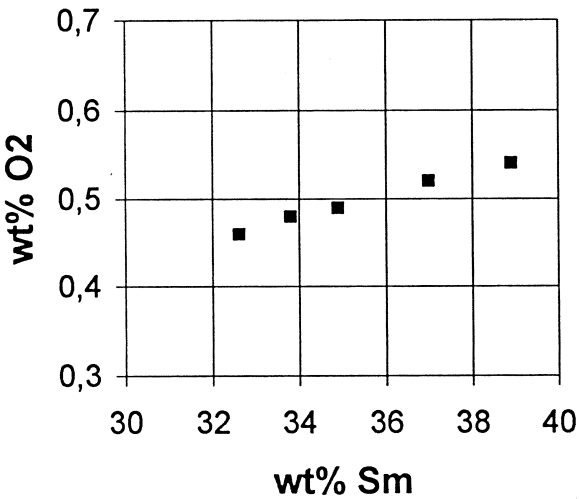

Fig. 3. Oxygen content of samples mechanically alloyed for 3000 mins and heat treated at 875°C for 2 hours.

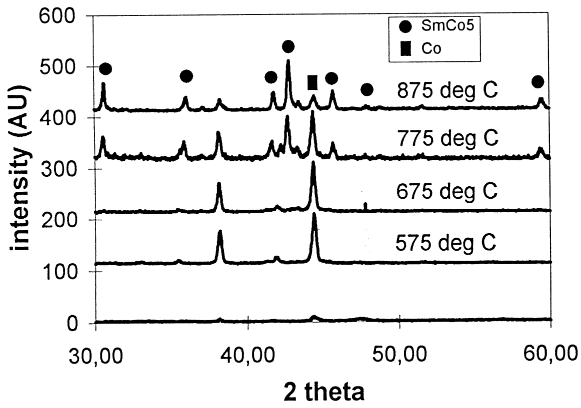

Fig. 4. X-ray diffraction traces from samples of mechanically alloyed $Sm_{1.15}Co_5$ heat treated at 575°C, 675°C, 775°C and 875°C.

low intensity peaks characteristic of a mixture of nanocrystalline Co and a possible amorphous phase, the peaks at 2θ=475° and 44.5° can be indexed to Co (*P*63/*mmc*). Low temperature heat treatment at 575°C and 675°C, appears to develop an intermediate SmCo structure which has not been identified but at higher temperatures 775°C and 875°C there is good evidence for the $SmCo_5$ phase (*P*6/*mmm*) with all the strongest peaks from $d=$ 0.2929 nm to 0.1556 nm being accounted for. A significant amount of cobalt is retained during the development of the $SmCo_5$ phase at high temperatures, it is possible that this could result from oxidation during heat treatment. There is little evidence for the development of much 2:17 phase.

Prior to any heat treatment the as mechanically alloyed powder was found to be magnetically soft, and the saturation magnetisation was found to decrease with increasing amounts of samarium. The effect of the heat treatments on the room temperature magnetic properties is shown in Fig. 5. The graph shows the variation of coercivity, H_c, as a result of the different heat treatments. The low samarium content samples exhibit very poor coercivities of less than 1 kOe. Increasing the amount of samarium results in the development of substantial coercivity in the order of 8 kOe. Further increase in the amount of added Sm results in further increase in the materials resistance to demagnetisation. The largest coercivity, 9.8 kOe, was obtained with the $Sm_{1.25}Co_5$ alloy heat treated at 600–700°C. Coercivity was observed to be strongly linked to the Sm content of the powder.

In order to assess the degree of exchange-coupling taking place in these materials a graph of M_r/M_s vs heat treatment temperature was plotted, Fig. 6. The value of M_s being obtained from the first quadrant of the hysteresis loop. All the materials were observed to saturate and the curves exhibited a smooth hysteresis loop. The graph again shows very poor results for the low Sm materials $Sm_{0.95}Co_5$ and $Sm_{1.00}Co_5$ which have M_r/M_s values of only ~0.1. An increase in the Sm content to $Sm_{1.05}Co_5$ sees a very significant jump in the maximum value of M_r/M_s to ~0.69 for the material heat treated at 775°C. Similar although slightly lower values were obtained for the $Sm_{1.15}Co_5$ alloy. Increasing the samarium content further, leads to a decrease in the maximum value for M_r/M_s of ~0.5, i.e. half the value of the saturation polarisation and in accordance with that predicted from simple theory for an isotropic material [13].

Further studies into the material's structure and grain size will be undertaken in subsequent studies.

Ding et al. [10] carried out an investigation with SmCo powders somewhat less rich in Sm than those used in this series of experiments. In addition the enhanced remanence they report appears to come about as a result of exchange coupling between 2-17 and 1-5 phases, which is different to that observed in this study. Smith et al. [11] in their study of SmCoFe alloys again observed a different structure with the hard magnetic phase being a 1-7 or 2-7 phase and the soft phase being bcc FeCo. The differences in compositions make it hard to draw direct comparisons between these studies and the present one, however it does

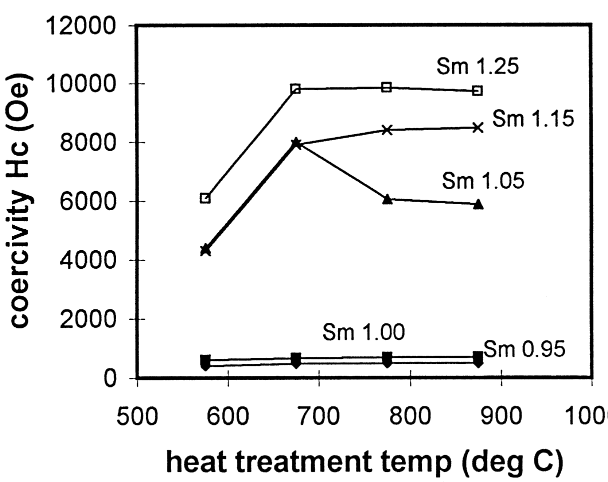

Fig. 5. The effect of heat treatment temperature on the coercivity of samples of Sm_xCo_5.

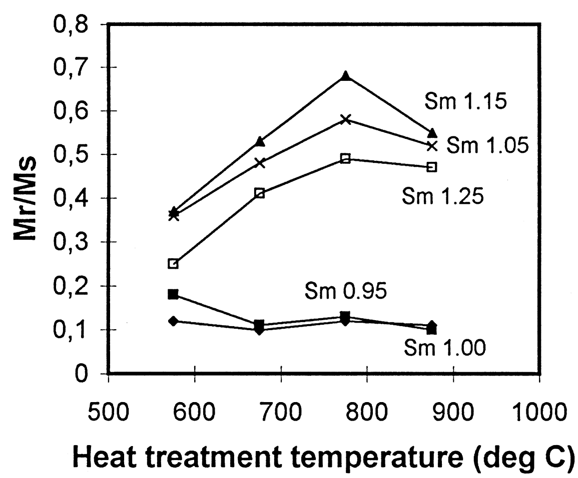

Fig. 6. The effect of heat treatment temperature on the ratio M_r/M_s.

seem clear that there are many possibilities for producing exchange coupled SmCo based materials over a range of compositions, mechanical alloying techniques and annealing temperatures.

4. Conclusion

Mechanical alloying of a range of SmCo powders with compositions in the range Sm_xCo_5 where $x=0.95$–1.25 produced amorphous material with soft magnetic properties. Mechanically alloyed particles were found to be basically spherical in shape. Heat treatments on these powders developed coercivities and a degree of remanence enhancement up to $M_r/M_s=0.69$.

Coercivity appears to develop as a consequence of the formation of the $SmCo_5$ phase and the remanence enhancement as a result of exchange-coupling between this $SmCo_5$ phase and cobalt.

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