# Magnetoresistance in Co-Cu metastable alloys prepared by mechanical alloying and shock-compression

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# Abstract

We had prepared a series of face-centered-cubic (FCC) structure supersaturated solid solution bulk alloys in cobalt (Co) - copper (Cu) (10-90 mol.% Co) system by mechanical alloying (MA) and shock compression. The magnetization and magnetoresistance (MR) were investigated for the solid solution alloys and the annealed alloys. The MR ratio increased with annealing temperature up to 400°C. The maximum MR ratio, under 10 kOe at room temperature, was 4.6% for the  $Co_{20}Cu_{80}$  sample annealed at 450°C for 20 minutes.

Key words: Co-Cu alloy, mechanical alloying, shock compression, magnetoresistance, magnetization

# 1. INTRODUCTION

Since the first observation of giant magnetoresistance (GMR) in Fe-Cr multilayer [1], a great deal of attention has been focused on this phenomenon. A GMR of greater than 50% at room temperature has been found in Co-Cu multilayer [2, 3]. In succession, a GMR of about 20% at low temperatures has been observed in a single layer film of Co-Cu alloy obtained by sputtering [4, 5]. Subsequently, the similar effect has been observed in various metastable granular Co-Cu alloys, in which nano-scaled Co-rich particles distributed in nonmagnetic Cu matrix, prepared by rapid liquid quenching, vapor deposition, electrodeposition and mechanical alloying (MA) methods [6-13]. It is now widely accepted that the MR effect in granular metals is attributed to the reorientation of the magnetic particle moments and spin dependent scattering of conduction electrons occurring in the magnetic particles as well as at the magnetic and nonmagnetic interface [14-16].

We had prepared Co-Cu system bulk alloys by MA combined with shock compression, and measured the magnetization at low temperature (4 K). The saturation magnetic moment at 0 K showed a fit curve to the Slater-Pauling one [17]. In this work, we investigated the MR in the solid solution bulk alloys as well as the annealed samples.

# 2. EXPERIMENTAL

Starting powders were provided by Rare Metallic Co., Ltd. The Co and Cu powders consisted of irregular particles of 1-2  $\mu$ m and 325 mesh (<44  $\mu$ m) in diameter, and the purity of Co and Cu in catalog were 99.9 and 99.99% wt.%, respectively. The Co concentrations ranged from 10 to 90 mol.%. The MA experiments were carried by using the planetary micro ball mill (P-7 of Fritsch Co., Ltd.) in an argon atmosphere grove box. A mill capsule and balls were used, which were made of silicon nitride  $(Si_3N_4)$  and zirconia  $(Y_2O_3$ -doped tetragonal ZrO<sub>2</sub>), respectively. The starting powder with a weight of 20 grams and 200 zirconia balls were contained into the capsule with a ball-to-powder weight ratio of about 4:1. The rotation speed of the ball mill was about 2840 rpm. The milling was interrupted each 30 min for 35 min to cool mill capsule to avoid heating. The milling duration was 21 hours, and small amount of the material were taken for analysis after selected milling times. Shock compression recovery experiments were performed using a propellant gun [18].

The structure of MA-treated and shock consolidated specimens were investigated by powder X-ray diffraction (XRD) and instrumental chemical analysis. Magnetization curves at room temperature were measured by using a vibrating sample magnetometer (VSM) device. Magnetoresistance (MR) was measured using a dc four probe method under magnetic field up to 10 kOe at room temperature. The rectangular bulk samples used in MR measurements had length, width and thickness of about 10 mm, 3 mm, and 1-2 mm, respectively, and were annealed in the temperature range of 250°C to 450°C for various times under vacuum. Annealing time is 20 minutes. MR ratio was calculated as the absolute value of  $\Delta \rho / \rho_0 = (\rho_H - \rho_0) / \rho_0$ , where  $\rho_0$  is the resistivity at zero field, and  $\rho_{\rm H}$  is the resistivity at the applied field of H.

# 3. RESULTS AND DISCUSSION

3.1 Metastable supersaturated solid solution alloys

The X-ray diffraction (XRD) patterns of the starting powder, MA-treated powder and the shock consolidated bulk body in  $Co_{50}Cu_{50}$  alloy are shown in Fig. 1. After 21 hours of milling, the Co peaks have fully disappeared, and only the broaden peaks of FCC phase can be observed. The XRD pattern of bulk body formed at an impact velocity of 1.0 km/s did not change much from that of the MA-treated powder. This showed that the



Fig. 1. X-ray diffraction patterns of the starting powder, the MA-treated powder, and the shock consolidated bulk body in  $Co_{50}Cu_{50}$  alloy.



Fig. 2. The lattice parameter versus Cu content of Co-Cu system MA-treated solid solution powder.

metastable solid solution powder was successfully consolidated without decomposition. The similar results were obtained for all other samples.

The lattice constants estimated from the diffraction peaks of FCC phase powders are shown in Fig. 2. The lattice parameters of 21 hours MA-treated powders were smaller than that of pure Cu, and larger than that of pure FCC-Co, respectively. It was found that the lattice parameter increases with Cu Concentration, but is greater than the data based on the Vegard's law, which means that the supersaturated solid solution was formed. The reason of the lattice parameter's change may be due to the magnetovolumetric effect. By using the Scherrer's equation the grain size was calculated to be 10-15 nm for 21 hours MA-treated powders. Shock consolidated bulk bodies have diameter and thickness of about 12mm and 2.5 mm, respectively. The relative density of shock consolidated bulk alloys approached about 90%. The chemical analytical result showed that

the average total impurity content in MA-treated powders and bulk alloys was less than 3 wt.%.

#### 3.2 Magnetoresistance

In order to obtain a large MR ratio annealing was performed on  $Co_{30}Cu_{70}$ ,  $Co_{20}Cu_{80}$ ,  $Co_{10}Cu_{90}$  bulk alloys in the temperature range of 250 to 450°C. Figure 3 shows the magnetic field dependence of MR ratio for  $Co_{20}Cu_{80}$  solid solution bulk alloy and the samples annealed at 250°C and 450°C. The MR ratio in  $Co_{20}Cu_{80}$  alloy annealed 450°C approaches 4.6% under 10 kOe, is about 4 times of the one in  $Co_{20}Cu_{80}$  alloy annealed at 250°C. It is worth mentioning that the MR ratio does not saturate under 10 kOe, and it may be more higher under a lager field. The magnetization also increases with the annealing temperature because the Co-rich particles gradually separate from the solid solution during annealing process.

Figure 4 shows the change of MR ratio obtained at room temperature under a magnetic field of 10 kOe with the annealing temperature for  $Co_{30}Cu_{70}$  alloy. The MR ratio increases with annealing temperature at first, and shows a maximum ratio of 4.2% after annealing at



Fig. 3. Magnetic field dependence of the MR ratio for  $Co_{20}Cu_{80}$  solid solution alloy and the samples annealed at 250°C and 450°C for 20 minutes.



Fig. 4. The change of MR ratio obtained at room temperature under 10 kOe with the annealing temperature.

400°C, then decreases. It is well known that the GMR or MR in magnetic granular metals is directly related to the magnetic particle size and their distribution in the nonmagnetic matrix, which directly determines the spin dependent scattering through the surface to volume ratio and particle distance. In particular, the value of MR increases initially with increasing of particle diameter and it decreases above a certain size. The maximum value occurs for the particle diameters around the electron mean free path. Therefore, it is easy to understand the change of MR ratio as above described.

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