High temperature magnetic properties of nanocrystalline $PrCo_5$ and YCo_5 alloys obtained by mechanical milling

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Abstract

Enhanced hard magnetic properties were obtained in nanostructured $PrCo_5$ and YCo_5 intermetallic compounds processed by mechanical milling for 240 min, and subsequent annealing in high vacuum at 1103 K for 1.0 and 2.5 min, respectively, followed by quenching in water. X-ray diffraction data demonstrate that the annealing has produced an average grain size <D> below 20 nm for both compounds, as a consequence magnetic measurements at room temperature showed a coercivity (HC) higher than 0.79 MA/m, and enhanced remanence ($\sigma_r / \sigma_{max} > 0.5$). High temperature magnetic measurements showed a temperature dependence of H_C and remanent magnetization σ_r for both compounds, where the temperature coefficient of coercivity for $PrCo_5$ is bigger than for YCo_5. Such behavior is consistent with the intrinsic temperature variation of magnetocrystalline anisotropy for $PrCo_5$ and YCo_5 intermetallic compounds.

Introduction

Since their discovery in the late 1960s, rare-earth-cobalt based magnetic materials have attracted considerable attention due to their large anisotropy fields H_A, relatively high saturation magnetizations M_s, and high Curie temperatures T_C.[1] In the past years the interest in this kind of materials has been increased due to their superior high temperature properties.[2] Most of the interest on this type of alloys has been focused on $SmCo_5$ due its exceptionally high H_A that facilitates high coercivity H_c, which is combined with a relatively high maximum energy product $(BH)_{max}$, an important figure of merit for permanent magnets.[2] Since this phase has been processed by different techniques,[3–6] other compounds in rare-earth-cobalt based family have attracted attention for their good magnetic properties, including $PrCo_5$ and YCo_5. High coercivities have been obtained for mechanically milled YCo_5 and $PrCo_5$ with a nanoscale grain size, stimulating interest on these materials for possible permanent magnet applications.[7–9] Likewise, from the strong research that has been done to improve the coercivity at high temperature for permanent magnets, the most promising compositions are based on rare-earth and transition metal alloys, with a 1:5-based composition. In this paper, we report the microstructure and the high temperature magnetic properties of nanostructured $PrCo_5$ and YCo_5 intermetallic compounds obtained by the mechanical milling technique.

Experimental

The alloys with nominal composition $Pr_{1.07}Co_5$ and $Y_{1.07}Co_5$ were prepared by arc melting pure elements in an Ar atmosphere. Afterwards, the as-cast ingots were coarsely pulverized, and the powders were mechanically milled for 240 min. The milling was carried out under Ar atmosphere by using a SPEX 8000 ball mill with a powder to ball's ratio of 1:8. The as-milled amorphous powder were annealed at 1103 K for 1 min for $PrCo_5$ and 2.5 min for YCo_5; in each case it was carried out in high vacuum closed Vycor ampoules and followed by quenching in water. X-ray diffraction (XRD) analysis was performed on finely ground powder with an automated Siemens model D5000 diffractometer with graphite monochromator (Cu Kα radiation). Hysteresis loops were measured with an Oxford Maglab 9100 vibrating sample magnetometer for temperature ranging from 300 to 923 K, applying a maximum field of 3.98 MA/m.

Results and discussion

Figure 1 shows the XRD patterns of $PrCo_5$ and YCo_5 after 240 min of mechanical milling and subsequent annealing at 1103 K for 1 and 2.5 min, respectively, followed by quenching in water. All the diffraction peaks could be indexed with the $CaCu_5$ hexagonal structure (PDF-2, 1996). Peaks' broadening is due to the reduced crystallite size of $PrCo_5$ and YCo_5 grains. The asymmetry in the (110) and (002) peaks for both 1:5 phases suggests a small quantity of 2:17 secondary phase in both cases, with a more evident presence in $PrCo_5$, but calculations made with X'Pert High- Score Plus software from PANalytical confirm that the 1:5 phase represents more than 90 wt % of the annealed powder for both samples. The cell parameter values, determined with the UNITCELL program,[10] and average grain size <D> are presented in Table I. These values are in good agreement with reported values for stoichiometric PrCo5 and YCo_5.[7,11] Average grain size <D> values are well within the range required for strong exchange coupling between adjacent crystallites.[12]

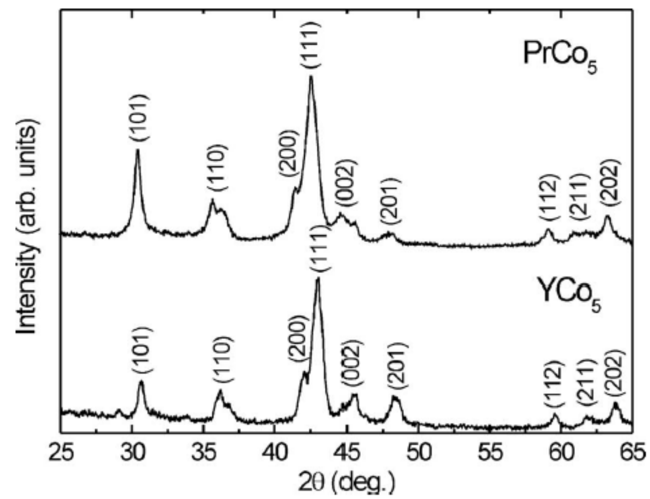

FIG. 1. XRD patterns for $PrCo_5$ and YCo_5 powders after milling for 240 min followed by a annealing at 1103 K by 1 and 2.5 min, respectively.

Figure 2 shows the room temperature hysteresis loops for nanocrystalline $PrCo_5$ and YCo_5. Nanocrystalline $PrCo_5$ intermetallic compound showed the highest coercivity H_C (=1.34 MA/m) combined with enhanced remanence σ_r/σ_{max}=0.66. Besides, nanocrystalline YCo_5 intermetallic compound showed a coercivity H_C value of 0.82 MA/m and an enhanced remanence ratio σ_r/σ_{max}=0.69. The initial magnetization curves for both compositions suggest a pinning type magnetization process,[13] in good agreement with that reported by Sánchez Ll. et al.[14] for the isostructural compound $Y_{0.5}Pr_{0.5}Co_5$. Due to the very fine average grain sizes below 20 nm for those materials, the intercrystallite exchange coupling penetrates deeply in the nanocrystallite volume and gives rise to the formation of multicrystallite or interaction domains, where the possible pinning sites must be located at the crystallite boundaries.[15]

Figure 3(a) shows the coercivity H_C of nanostructured $PrCo_5$ and YCo_5 intermetallic compounds measured as a function of temperature; in both cases coercivity decreases as a result of the temperature increment. For nanostructured

$PrCo_5$, coercivity decreases from 1.34 MA/m at room temperature to 0.13 MA/m at 723 K, whereas the respective values for YCo5 are 0.82 and 0.20 MA/m. It can be seen that H_C drops very steeply for the nanocrystalline $PrCo_5$ powder, while the corresponding curve for YCo_5 exhibits weaker temperature dependence. The convergent behavior of the curves shows the high temperature sensitivity of Pr anisotropy, due to the weak Pr–Co intersublattice exchange that is easily overcome by thermal excitation.[16] Remanent magnetization dependence on temperature for nanostructured $PrCo_5$ and YCo_5 powders is shown in Fig. 3(b), where remanent magnetization values for nanocrystalline YCo_5 exposed a higher temperature dependence than for nanocrystalline $PrCo_5$ compound.

TABLE I. Cell parameters and average crystallite size $\langle D \rangle$ of $PrCo_5$ and YCo_5 intermetallic alloys after mechanical milling and heat treatment.

Composition	*a* (Å)	*c* (Å)	$\langle D \rangle$ (nm)
YCo_5	5.027 ± 0.001	3.989 ± 0.001	15.0 ± 0.1
$PrCo_5$	4.950 ± 0.001	3.973 ± 0.001	17.9 ± 0.1

The observed decreasing of σ_r is due to magnetic dealignment between the magnetic moment of adjacent nanograins, which is originated from the enhanced thermal energy as the temperature increases. Both σ_r versus T curves showed a very similar behavior in all temperature range, reaching the lower σ_r value at 920 K, which is very close the Curie temperature T_C for both compounds, with values of 940 and 980 K for $PrCo_5$ and YCo_5, respectively.[17,18]

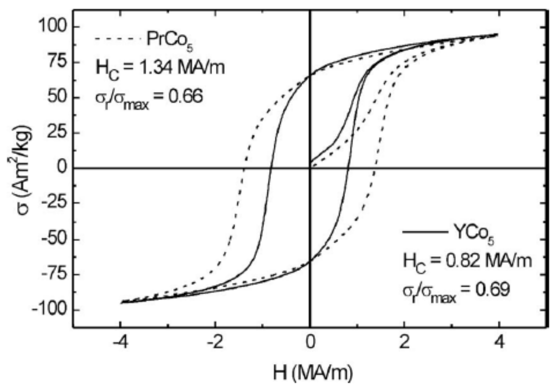

FIG. 2. Room temperature hysteresis loops of nanocrystalline $PrCo_5$ and YCo_5 powders after mechanical milling and heat treatments at 1103 K for 1 and 2.5 min, respectively.

Table II shows the temperature coefficients of coercivity and the remanent magnetization for the nanostructured $PrCo_5$ and YCo_5 intermetallic compounds. From 300 to 573 K, the temperature coefficient of remanent magnetization α is more negative for nanocrystalline $PrCo_5$. On the contrary, from 573 to 873 K, α is more negative for YCo_5. In addition, the temperature coefficient of coercivity β is more negative for $PrCo_5$ than for YCo_5 for lower temperature values, due to the stronger dependence of Pr magnetocrystalline anisotropy with temperature than for Co. Finally, since coercivity in the YCo_5 phase is only dependent on the cobalt sublattice anisotropy,[16,19] there is an almost linear behavior of β in all the temperature ranges. The magnetic behavior observed in these nanostructured $PrCo_5$ and YCo_5 powders is consistent with the temperature variation of magnetocrystalline anisotropy for both intermetallic compounds.[16]

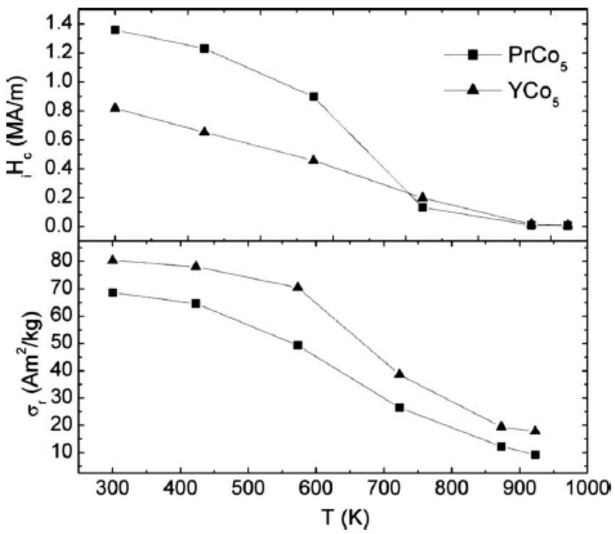

FIG. 3. Coercivity H_C and remanent magnetization σ_r dependence on temperature for nanocrystalline $PrCo_5$ and YCo_5 powders, with T ranging from room temperature to 923 K.

TABLE II. Temperature coefficients of coercivity β and remanent magnetization α for the nanostructured $PrCo_5$ and YCo_5 powders.

Composition	Temperature range (K)	α (%/K)	Temperature range (K)	β (%/K)
YCo_5		-0.036 ± 0.009		-0.176 ± 0.008
$PrCo_5$	300–573	-0.072 ± 0.019	300–723	-0.209 ± 0.047
YCo_5		-0.148 ± 0.019		-0.148 ± 0.002
$PrCo_5$	573–873	-0.114 ± 0.012	723–873	-0.060 ± 0.002

Conclusions

Enhanced hard magnetic properties were obtained in nanostructured $PrCo_5$ and YCo_5 intermetallic compounds processed by mechanically milling for 240 min, annealed in high vacuum at 1103 K for 1.0 and 2.5 min, respectively, and subsequently quenched in water. Magnetic measurements of both compounds have shown their largest magnetic properties at room temperature. XRD data demonstrate that the annealing time t has produced an average grain size <D> below 20 nm for both compounds. High temperature magnetic measurements showed that the temperature coefficient of coercivity for $PrCo_5$ is bigger than for YCo_5. Such behavior is in good agreement with the intrinsic temperature dependence of magnetocrystalline anisotropy for $PrCo_5$ and YCo_5 intermetallic compounds.

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