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The effect of manganese substitution on the magnetic properties of $\text{Ce}_2\text{Co}_{17}$ compounds

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Abstract. X-ray diffraction (XRD) and magnetization measurements were employed to investigate the effect of Mn substitution for Co on the structure and magnetic properties of $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ compounds with $x = 0, 1, 2, 3, 4$. XRD patterns show that all samples are single phase with the hexagonal $\text{Th}_2\text{Ni}_{17}$ -type structure. The substitution of Mn for Co in $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ compounds leads to a monotonic increase in unit-cell volume. The saturation magnetization at 5 K and the Curie temperature T_C decrease monotonically with increasing Mn content. X-ray diffraction measurements on magnetically aligned $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ powders with $x = 0-4$ exhibit an easy-axis type of magnetic anisotropy at room temperature. The substitution of Mn for Co leads to a change of the magnetocrystalline anisotropy of the Co sublattice from the basal plane to c -axis and strongly increases the uniaxial anisotropy of $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ compounds. The magnetocrystalline anisotropy field H_A is found first to increase and then decrease with increasing Mn concentration, attaining a maximum value of 30.1 kOe at $x = 2$.

1. Introduction

Intermetallic compounds based on rare-earth elements (R) and 3d-transition elements (T) form a large class of materials that are important from a technological as well as from a fundamental point of view. Rare-earth cobalt intermetallic compounds R_2Co_{17} have attracted much attention since the discovery of $\text{Sm}_2\text{Co}_{17}$ magnets. Although they have high saturation magnetization, it is hard to find practical applications for the R_2Co_{17} series compounds in high-energy product magnets, due to their unfavourable anisotropy. These compounds have either weak uniaxial (e.g., $\text{Sm}_2\text{Co}_{17}$) or planar (e.g., Y_2Co_{17} , $\text{Pr}_2\text{Co}_{17}$) anisotropy. In previous investigations, it was found that substitution of non-magnetic atom like Ga, Al, or Si for Fe in R_2Fe_{17} compounds has an important influence on the formation, structure and magnetic properties, especially the magnetocrystalline anisotropy. A change of the magnetocrystalline anisotropy with Ga or Al content from planar to axial was observed in $\text{R}_2\text{Fe}_{17-x}\text{M}_x$ with $\text{M} = \text{Ga}$ or Al at room temperature [1, 2]. Recent studies have shown that the substitution of nonmagnetic atoms Ga, Si and Al also has a significant effect on the magnetocrystalline anisotropy in R_2Co_{17} compounds [3–12]. Among all $\text{R}_2\text{Co}_{17-x}\text{M}_x$ compounds, a variety of anomalous behaviours in the lattice constants and magnetic properties were observed for Ce compounds [6–9] due to the mixed-valence behaviour of the Ce ion, which has attracted much interest recently [13]. It is well known that

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there is a close connection between the valence and the magnetic character of cerium. In intermetallic compounds with transition metals, cerium is more often found in the nonmagnetic and strongly mixed-valent α state, than in the magnetic trivalent γ state. The Cerium 4f wavefunction is strongly hybridized with the (5d6s) wavefunction in the α state. In the γ state, the 4f wavefunction is almost atomlike and produces a large magnetic anisotropy; the cerium magnetic moment is close to $2.54 \mu_B$, the theoretical magnetic moment of trivalent cerium.

In this paper, the effect of Mn substitution for Co in $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ ($x = 0-4$) compounds on the structure and magnetic properties is reported.

2. Experiment

The $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ ($x = 0-4$) alloys were prepared by arc melting the constituent metals in an argon atmosphere of high purity. The raw materials of Ce, Co, Mn were at least 99.9% pure. The as-cast ingots were annealed under vacuum at 1273 K for 100 h, then quenched into water. Powder x-ray diffraction using Cu $K\alpha$ radiation was used to determine phase purity, crystal structure and lattice parameters. The saturation magnetic moment μ_s at 5 K was deduced from the data collected on free powder samples using a SQUID magnetometer in a field up to 6.5 T. The Curie temperature T_C was determined from the temperature dependence of magnetization measured with a Faraday balance in a field of 0.1 T for 300–1200 K (± 1 K). The aligned samples for the magnetocrystalline anisotropy studies were prepared by mixing the well ground fine powder with epoxy resin and then aligning in a magnetic field of 1 T. The x-ray diffraction procedure was also used to determine the easy magnetization direction at room temperature. The anisotropy fields were obtained from the magnetization curves measured along and perpendicular to the direction of alignment.

3. Results and discussion

X-ray diffraction patterns indicate that all samples are single phase with the hexagonal $\text{Th}_2\text{Ni}_{17}$ -type structure. Figure 1 shows examples of the x-ray diffraction patterns of $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ compounds with $x = 1$ and 4. It is found that $\text{Ce}_2\text{Co}_{17-x}\text{M}_x$ ($M = \text{Ga}, \text{Al}, \text{Si}$) [6–9] have the hexagonal $\text{Th}_2\text{Ni}_{17}$ -type structure at low substitution amount and the rhombohedral $\text{Th}_2\text{Zn}_{17}$ -type structure for high M concentration. The previous studies have shown that the R_2Co_{17} compounds crystallize in two different structure types [14]. The compounds in which the R component has a comparatively large metallic radius adopt the rhombohedral $\text{Th}_2\text{Zn}_{17}$ structure. The hexagonal $\text{Th}_2\text{Ni}_{17}$ structure is found for R elements of smaller metallic radius, starting from R = Gd when moving across the rare-earth series. In fact, $\text{Gd}_2\text{Co}_{17}$ is at the border of the two structural regions, and this is most likely the reason why it can form in either one of the two structure types, depending on the preparation conditions. $\text{Ce}_2\text{Co}_{17}$ shows a similar behaviour [15], which is apparently due to the fact that Ce is not trivalent in this compound and then its metallic radius is more like that of the R elements in the middle of the rare-earth series. The room-temperature lattice parameters a , c , c/a and the unit-cell volumes v are listed in table 1. It can be found that the c/a ratio increases with increasing Mn content, which may be due to the preferred substitution of Mn into some crystallographic sites. The unit cell volume is found to increase nearly linearly at a rate of about 2.95 \AA^3 per Mn atom, as shown in figure 2. A similar increase of unit cell has been also observed in $\text{R}_2\text{Co}_{17-x}\text{Al}_x$ [6, 7, 9, 10] compounds. The unit-cell volume of the $\text{Ce}_2\text{Co}_{17-x}\text{M}_x$ compounds is smaller than that of the corresponding Pr compounds [11]. This can be explained by the fact that Ce remains tetravalent or mixed valent as it does in $\text{Ce}_2\text{Co}_{17-x}\text{Al}_x$ compounds [9].

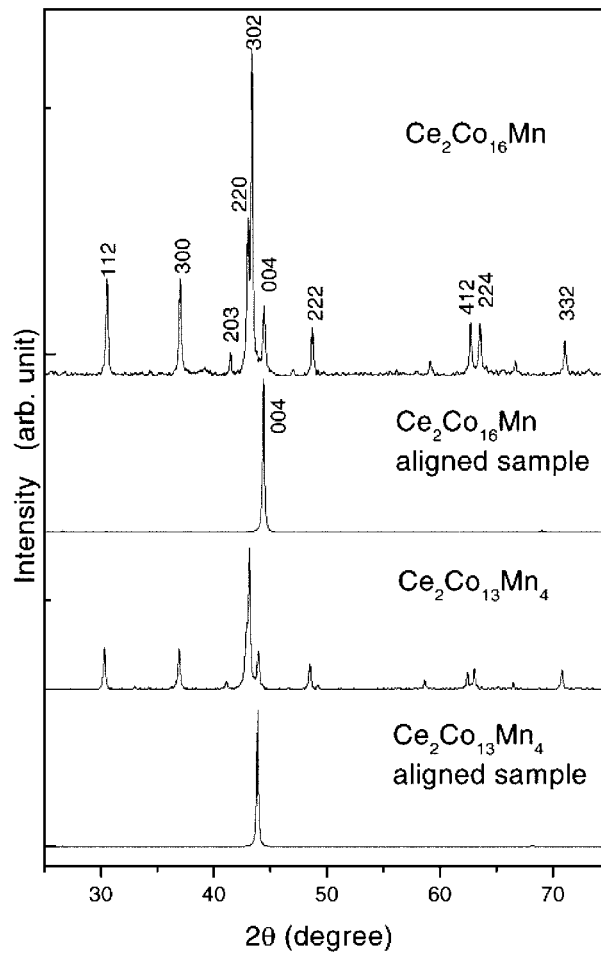


Figure 1. X-ray diffraction patterns at room temperature of powder and aligned samples of $Ce_2Co_{17-x}Mn_x$ compounds with $x = 1, 4$.

Table 1. The lattice parameters a , c , c/a , unit-cell volume v and crystalline structure type of $Ce_2Co_{17-x}Mn_x$ compounds.

Compounds	a (Å)	c (Å)	c/a	v (Å ³)	Structure
Ce_2Co_{17}	8.407	8.130	0.967	495.1 ± 0.1	Hexagonal
$Ce_2Co_{16}Mn$	8.409	8.143	0.968	498.6 ± 0.1	Hexagonal
$Ce_2Co_{15}Mn_2$	8.416	8.173	0.971	501.3 ± 0.1	Hexagonal
$Ce_2Co_{14}Mn_3$	8.422	8.213	0.975	504.5 ± 0.1	Hexagonal
$Ce_2Co_{13}Mn_4$	8.430	8.237	0.977	506.9 ± 0.1	Hexagonal

Figure 3 presents the magnetization curves of $Ce_2Co_{17-x}Mn_x$ free powders at 5 K. The saturation moment μ_s was obtained from fitting the data of $M(H)$ versus H by using the law of approach to saturation and is listed in table 2. The concentration dependence of the saturation magnetization M_s of the $Ce_2Co_{17-x}Mn_x$ compounds is also shown in figure 4. The saturation magnetization μ_s decreases slowly for $x \leq 2$, then decreases quickly with increasing Mn

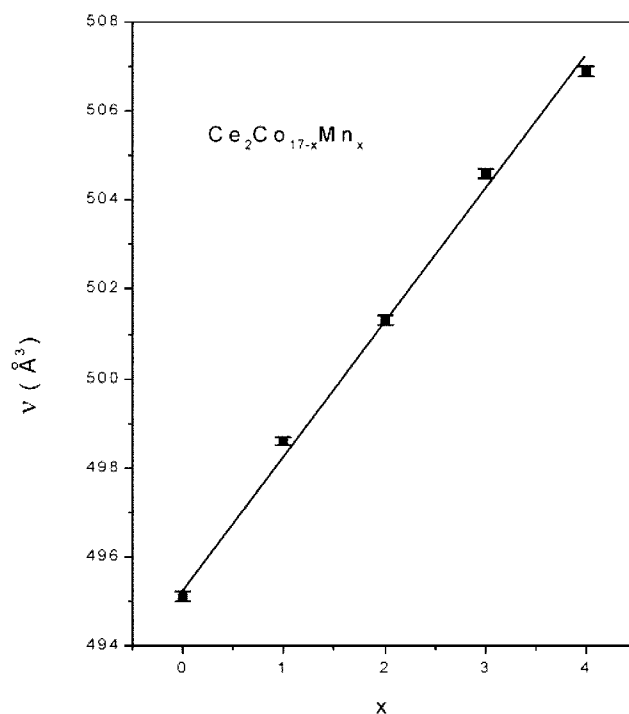


Figure 2. The unit cell volumes v as a function of Mn concentration x in $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ compounds. The solid line is plotted as a guideline for the eyes.

content. This behaviour is quite different from that of $\text{Ce}_2\text{Co}_{17-x}\text{M}_x$ ($M = \text{Ga}, \text{Al}, \text{Si}$) which shows a nearly linear decrease of saturation magnetization [6]. Among these compounds ($M = \text{Ga}, \text{Al}, \text{Si}, \text{Mn}$), the Si atom has strongest effect on the saturation magnetization, while the Mn atom has the weakest effect. Also the declines in saturation magnetization of $\text{Ce}_2\text{Co}_{17-x}\text{M}_x$ ($M = \text{Ga}, \text{Al}, \text{Si}$) are faster than that in the case of a simple magnetic dilution. From figure 4, the μ_s of $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ is larger than the value of the simple dilution before $x = 2.5$, while μ_s is lower than that of the simple dilution for $x > 2.5$. This phenomenon implies that the change in saturation magnetization is not only due to the reduced Co content, but also due to the change of the magnetic moment per Co atom. It is well known that the large spatial extent of the 3d wavefunctions leads to 3d-electron energy bands rather than to a 3d level [16]. The substitution of M not only changes the 3d-band structure, but also transfers the outer electrons to the 3d band. Merely from the changes of the unit cell volume, the width of the 3d band is different for different substitution $M = \text{Si}, \text{Al}, \text{Ga}, \text{Mn}$. This could explain the different results of magnetization for different substitution M.

In the compounds containing light rare-earth ions, the R moments and the Co moments are ferromagnetically coupled and the saturation moment μ_s can be expressed by $\mu_s = 15\mu_{\text{Co}} + 2\mu_{\text{R}}$, where μ_{Co} and μ_{R} are the average Co moment and the R moment, respectively. If we assume that the average Co moment is the same as that of the $\text{Y}_2\text{Co}_{17-x}\text{Mn}_x$ compounds [12] for Y has no magnetic moment, we can derive the Ce moments μ_{Ce} from the saturation moment μ_s , as shown in table 2. It can be seen that μ_{Ce} is negative and the absolute value increases with the Mn content. In the previous studies [17, 18], it was also found that the magnetic moment of Ce compounds is anomalously low when compared to Y and Lu compounds. This may

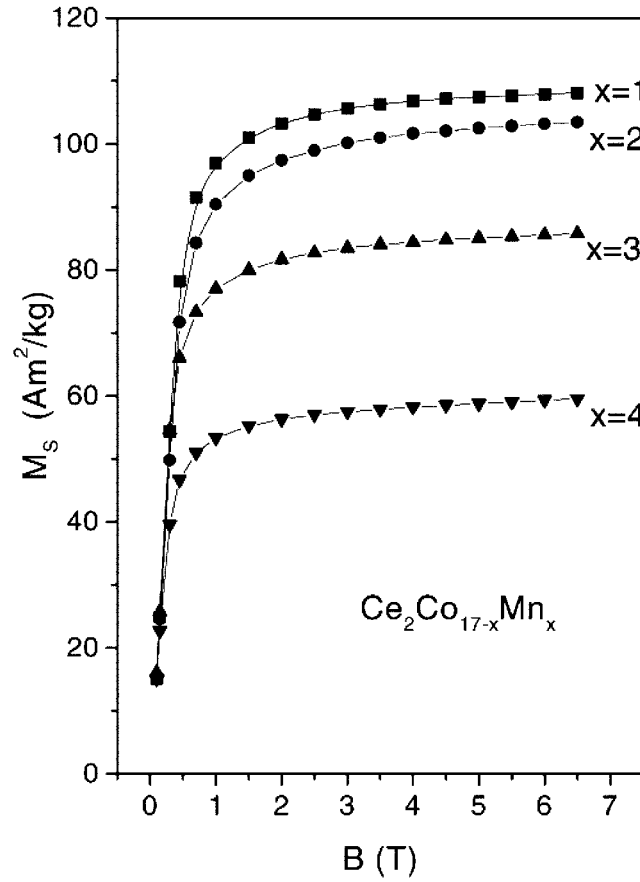


Figure 3. Field dependence of the magnetization at 5 K of $Ce_2Co_{17-x}Mn_x$ compounds measured on free powders.

Table 2. The saturation magnetic moment μ_s at 5 K, saturation magnetic moment μ_Y of $Y_2Co_{17-x}Mn_x$, the magnetic moment of Ce ion μ_{Ce} , Curie temperature T_C , easy magnetization direction (EMD) at room temperature (RT) and magnetocrystalline anisotropy field H_A of $Ce_2Co_{17-x}Mn_x$ compounds. For μ_Y the values are taken from [12], and μ_{Ce} is derived from μ_s and μ_Y according to the formula given in the text.

Compounds	μ_s ($\mu_B \text{ fu}^{-1}$)	μ_Y ($\mu_B \text{ fu}^{-1}$)	μ_{Ce} (μ_B)	T_C (K)	EMD at RT	H_A (kOe)
Ce_2Co_{17}	26.1 ± 0.1	28.0 ± 0.1	-0.95	1100 ± 1	<i>c</i> -axis	7 ± 1
$Ce_2Co_{16}Mn$	25.3 ± 0.1	27.7 ± 0.1	-1.2	927 ± 1	<i>c</i> -axis	29 ± 1
$Ce_2Co_{15}Mn_2$	24.3 ± 0.1	28.0 ± 0.1	-1.85	772 ± 1	<i>c</i> -axis	30 ± 1
$Ce_2Co_{14}Mn_3$	20.0 ± 0.1	27.5 ± 0.1	-3.75	526 ± 1	<i>c</i> -axis	25 ± 1
$Ce_2Co_{13}Mn_4$	14.1 ± 0.1	25.3 ± 0.1	-5.6	350 ± 1	<i>c</i> -axis	10 ± 1

be further evidence that Ce is not in the magnetic trivalent γ state with the magnetic moment $2.54 \mu_B$, the theoretical magnetic moment of trivalent cerium. From the calculated electronic structure of $CeCo_5$ [19], it is found that the anomalous properties of $CeCo_5$ are due to an itinerant 4f state which is caused by the hybridization of the cerium 4f states and the cobalt 3d

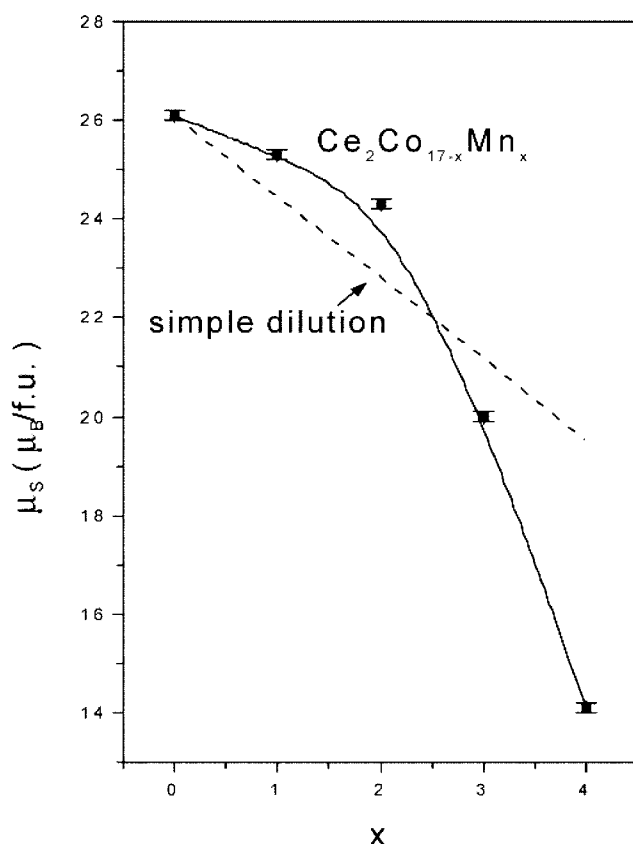


Figure 4. The Mn concentration dependence of saturation magnetization at 5 K. The simple magnetic dilution is also plotted as a dashed line. The solid line is plotted as a guideline for the eyes.

states [20]. So the Ce ion may have a strong effect on the magnetic moment of Co, thus the assumption that the average Co moment is the same as that of the $\text{Y}_2\text{Co}_{17-x}\text{Mn}_x$ compounds is not appropriate. Until now there has been little direct evidence on the valence state of $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ compounds, but the x-ray-absorption near-edge structure (XANES) spectra obtained at the cerium L-III edge of the $\text{Ce}_2\text{Fe}_{17-x}\text{Al}_x$ solid solutions indicate that cerium is in a mixed-valent state in these compounds. The cerium spectroscopic valence obtained from the relative areas of the two peaks decreases from 3.64 to 3.43 between $x = 0$ and 9 [13]. Also it is observed that the unit-cell volume, Curie temperature and saturation magnetization of $\text{Ce}_2\text{Co}_{17-x}\text{Al}_x$ compounds [9] are anomalously low in comparison with the values obtained from a simple extrapolation for the other $\text{R}_2\text{Co}_{17-x}\text{Al}_x$ compounds, such as $\text{R} = \text{Pr}$ and Nd . These anomalies also demonstrate mixed-valence behaviour of the Ce ion.

Figure 5 gives the temperature dependence of magnetization of $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ ($x = 0-4$) compounds. The Curie temperature is also listed in table 2 and is shown in figure 6 as a function of Mn concentration. From figure 6 it is found that Curie temperature T_C decreases monotonically with increasing Mn concentration. The decrease in T_C is estimated as 190 K per Co atom replaced by Mn in $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ compounds. It is well known that the Curie temperature is mainly determined by the T-T interaction in the rare-earth transition metal

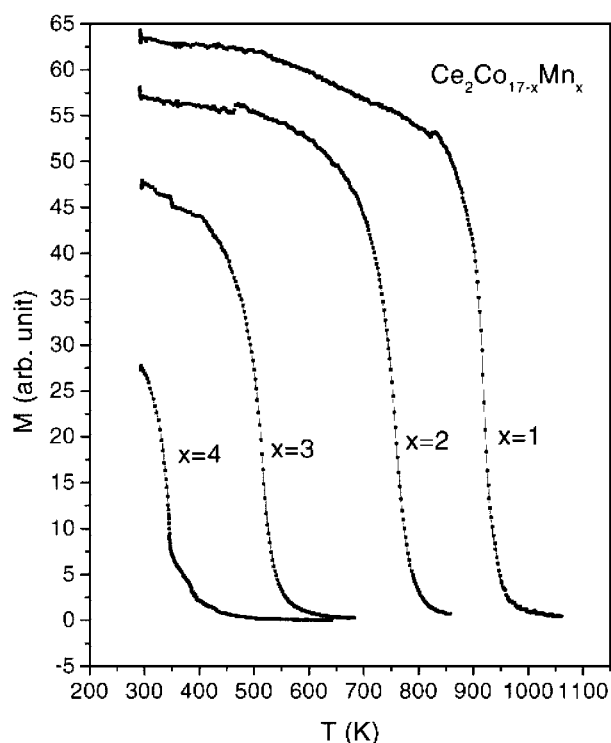


Figure 5. The temperature dependence of the magnetization of $Ce_2Co_{17-x}Mn_x$ compounds.

intermetallic compounds. In the molecular-field description, if we only consider the T-T interaction, the relation between T_C and exchange interaction constant should be:

$$3k_B T_C = 2J_{TT} Z_{TT} S_T (S_T + 1).$$

Here, J_{TT} stands for the T-T exchange interaction constant. S_T is the so-called pseudospin, which is related to the magnetization of the 3d sublattice M_T by:

$$M_T = -g_T S_T \mu_B.$$

So T_C is determined both by J_{TT} and M_T . Both the decreases of Co-Co exchange interaction and magnetization result in a rapid decrease of T_C with increasing Mn concentration. This situation is dramatically different from that observed in $R_2Fe_{17-x}Mn_x$ compounds where the Curie temperature first increases with x before decreasing at higher x value [21]. This may be due to the existence of antiferromagnetic coupling between Fe moments on certain sites: the preferential substitution of Mn on these sites can reduce the antiferromagnetic coupling and consequently enhance the T_C , while the magnetic coupling between Co moments in the metal system is always ferromagnetic, independent of Co moment and structure. Also the Curie temperature for $Ce_2Co_{17-x}Mn_x$ compounds is anomalously low in comparison with that of $Y_2Co_{17-x}Mn_x$ compounds with the same Mn concentration. This anomaly may be further evidence of the mixed-valence behaviour of the Ce ion.

X-ray diffraction measurements on magnetically aligned powder samples demonstrate that the easy magnetization direction (EMD) of $Ce_2Co_{17-x}Mn_x$ compounds is c -axis for $x = 0-4$. The EMD at room temperature is listed in table 2. Figure 1 also illustrates the XRD pattern of the magnetically aligned sample with $x = 1$ and 4 as an example. The drastic increase of

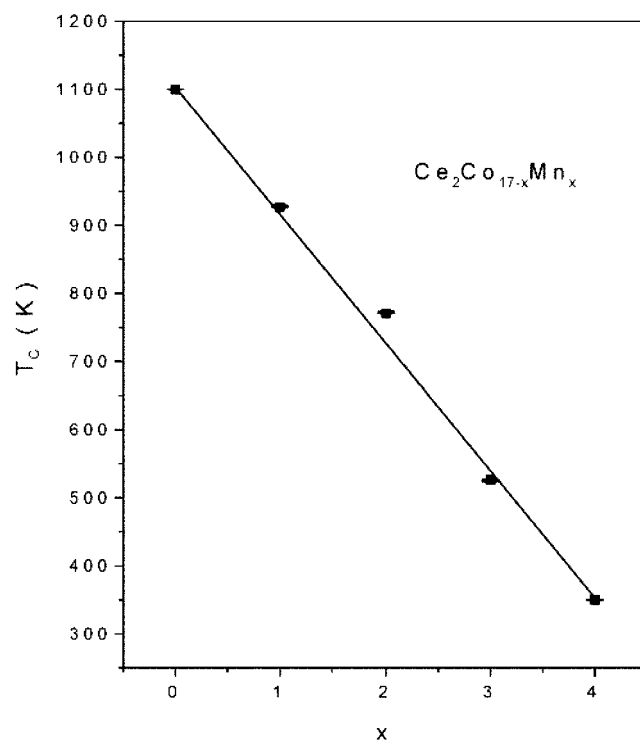


Figure 6. The Mn concentration dependence of the Curie temperature of $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$ compounds. The solid line is plotted as a guideline for the eyes.

the (004) peak and the complete disappearance of the ($hk0$) reflection reveal that the samples possess uniaxial anisotropy at room temperature. For $\text{Ce}_2\text{Co}_{17}$, the result of the uniaxial anisotropy has been also found in the single-crystalline [18] and polycrystalline samples of $\text{Ce}_2\text{Co}_{17}$ [20]. Previous investigations have shown that the anisotropy of the Co sublattice in R_2Co_{17} , as derived from studying Y_2Co_{17} , $\text{Gd}_2\text{Co}_{17}$ and $\text{Lu}_2\text{Co}_{17}$, favours the basal plane in the temperature range below at least 900 K [22]. This means that the contribution to the bulk anisotropy from the Ce ions of $\text{Ce}_2\text{Co}_{17}$ is uniaxial. The c -axis anisotropy of the Ce sublattice overcomes the relatively weak planar anisotropy of the Co sublattice over the whole magnetically ordered temperature range. The origin of the c -axis anisotropy of the Ce sublattice may be the hybridization of the Ce 4f states and the Co 3d states [20]. The overall anisotropy of R_2Co_{17} compounds is determined from the sum of the Co sublattice anisotropy and the rare-earth sublattice anisotropy. It has been shown from the study of $\text{Y}_2\text{Co}_{17-x}\text{Mn}_x$ compounds [12] that the Mn atom plays an important role in determining the easy magnetization direction in these compounds. The substitution of Mn for Co leads to a decrease in the planar anisotropy of the Co sublattice and then changes the magnetic anisotropy from the basal plane to the c -axis at room temperature.

Figure 7 presents the concentration dependence of the magnetocrystalline anisotropy field H_A at 300 K. The anisotropy field was obtained by an extrapolation procedure from the magnetization curves measured along and perpendicular to the aligned direction. H_A is found first to increase and then decrease with increasing Mn concentration, attaining a maximum value of 30.1 kOe at $x = 2$. Such behaviour can also be found in other $\text{Ce}_2\text{Co}_{17-x}\text{Mn}_x$

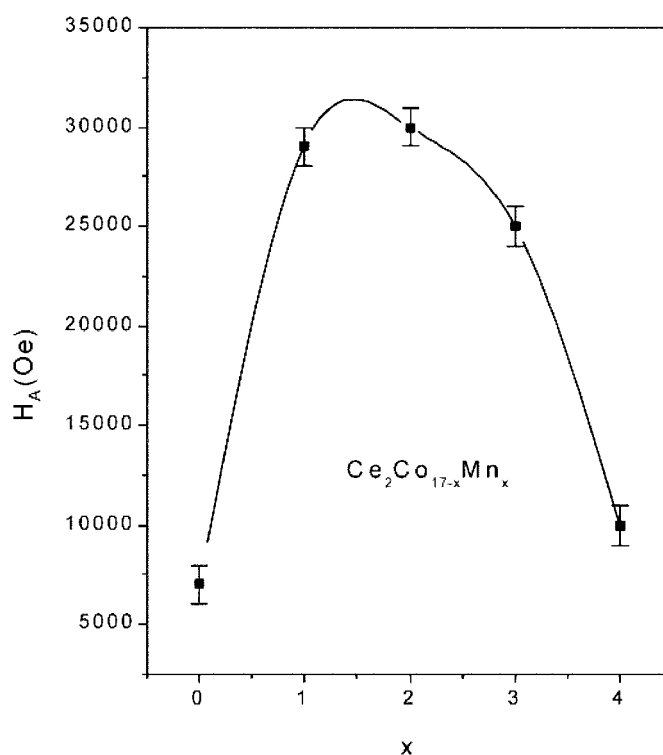


Figure 7. Magnetocrystalline anisotropy field H_A at room temperature as a function of Mn concentration. The solid line is plotted as a guideline for the eyes.

compounds [6–9]. The enhancement of H_A at low Mn concentration is related to the change of magnetic anisotropy of the Co sublattice from planar to axial in $Ce_2Co_{17-x}Mn_x$ compounds by the preferential substitution of Mn. In the previous study, the change of the easy magnetization direction was attributed to a preferential substitution of the alloying elements (Al, Ti, Mn, Fe) for the plane seeking 6c dumbbell cobalt sites [11, 23]. Inomata [24] has proposed that the 18f site of Co in Ce_2Co_{17} makes a positive contributions to the anisotropy (c -axis), while the 18h, 9d and 6c sites make negative contributions (basal plane). In particular, the 6c site was the largest negative contribution. The preferential substitution of Mn into ‘dumb-bell’ pairs found support in results of NMR investigation of these systems [25]. More recently, the neutron diffraction study on $Tb_2Co_{17-x}Ga_x$ compounds [26] demonstrated that Ga prefers to occupy the 18h sites, which also increases the c -axis anisotropy of the Co sublattice. At high Mn content, the strong decrease of Co moment also affects the anisotropy and results in a decrease of anisotropy.

Acknowledgments

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