

## STOICHIOMETRIC STUDIES ON EFFECT OF CO ON THE MAGNETIC PROPERTIES OF $\text{YGdFe}_{17}\text{Si}$

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**Abstract:** The structural and magnetic properties of the off-stoichiometric  $\text{YGdFe}_{17-x}\text{Co}_x\text{Si}$  ( $x = 0, 1, 2$  and  $3$ ) have been investigated through X-ray diffraction and magnetization studies. The lattice parameters, saturation magnetization and Curie temperature of the compounds were obtained. Structural characterization was carried out by taking X-ray diffraction patterns on sample powders. Magnetization and Curie temperatures were determined using a vibrating sample magnetometer up to an applied field of 12 kOe. All the compounds up to  $x = 3$  are formed in a single-phase  $\text{Th}_2\text{Ni}_{17}$  type hexagonal structure with the traces of  $\alpha$ -Fe. The unit cell volume decreased with the addition of Co due to the smaller size of Co than that of Fe. The saturation magnetization and Curie-temperature values are found to increase with the addition of Co from 130 emu/g to 150 emu/g and from 410 K to 708 K respectively. This could be due to the additional Co-Co, Fe-Co exchange whose magnitude is larger compared to that of Fe-Fe exchange. Magnetically aligned sample powders indicate the presence of planar isotropy for all the compounds.

*Keywords:* Curie-temperature, intermetallic, magnetization.

### Introduction

Rare earth-iron intermetallic compounds of the type  $\text{R}_2\text{Fe}_{17}$  ( $\text{R} = \text{rare earth}$ ) have attracted significant attention as possible candidates for high-performance permanent magnet materials [1]. These compounds have been reported to crystallize in the rhombohedral  $\text{Th}_2\text{Zn}_{17}$  type structure with the space group  $\text{R}\bar{3}\text{m}$ , for rare earths lighter than Gd and in the hexagonal  $\text{Th}_2\text{Ni}_{17}$  type structure with the space group  $\text{P}6_3/\text{mmc}$ , for rare earths heavier than Tb. The Curie temperatures ( $T_C$ ) of the  $\text{R}_2\text{Fe}_{17}$  compounds are low [ $\text{Gd}_2\text{Fe}_{17}$  is reported to have a  $T_C$  of 468 K, the highest]

due to the small exchange splitting in the 3d bands. In addition, the room temperature anisotropy which is predominantly due to the Fe sub lattice is small and planar. Partial substitution of non magnetic elements such as Al, Ga, and Si for iron has been reported to cause considerable increase in the  $T_C$  and magneto crystalline anisotropy of these compounds [2],[3]. These substitutional elements preferentially occupy 12j, 12k, and 6c (18h, 18f, and 6c) for Fe in hexagonal (rhombohedral) structure in the order of preference. Interstitial modification by N/C is known to improve the  $T_C$  as well as the magneto crystalline anisotropy in these compounds [4]. It has been reported that the 4p orbitals of Ga hybridize with the 3d band of Fe to form a common 4p, 3d band whereas Si and Al hybridize through (3p, 3d) hybridization. The partial density of states (PDOS) of Ga extends up to the Fermi level of the 3d band, and the PDOS of Si are to some extent near to the  $E_F$  whereas PDOS of Al lies at the bottom of the 3d band, which makes them alter the 3d band in a relatively less effective way resulting in smaller changes in the values of  $M_S$  and  $T_C$  in these compounds compared to the former ones [5], [6]. The magnetic properties of off-stoichiometric  $R_2Fe_{17}$  compounds of the type  $R_2Fe_{14}Si_2$  (TM-deficient  $R_2Fe_{17}$  type) have been investigated by Pourarian et al. [7] and  $R_2Fe_{17-x}Co_xSi$  (TM-Rich  $R_2Fe_{17}$  type) compounds by Ren et al [8]. They have reported that due to the dual (magnetic and nonmagnetic atoms) substitution, the enhancement in  $M_S$  and  $T_C$  in these compounds with substitution was larger compared to that of stoichiometric 2:17 compounds. As Al, Si, and Ga substitution for Fe cause an increase in the Curie temperatures of the  $R_2Fe_{17}$  compounds, it will be of interest to investigate the effect of addition of these elements on the magnetic properties. As Si causes the largest increase of the Curie temperatures, this has been chosen. Along with this, substitution of part of Fe by Co too was attempted as Co-based  $R_2Fe_{17}$  compounds are known to have high Curie temperatures. Thus, in this paper, the magnetic properties of the off stoichiometric  $YGdFe_{17-x}Co_xSi$  compounds are presented.

### Experimental details:

The  $YGdFe_{17-x}Co_xSi$  ( $x = 0 - 3$ ) compounds were prepared using an arc furnace and in an argon atmosphere, starting from Y and Gd of 99.9% purity, Fe and Si of 99.95% purity, and Co

of 99.99% purity. The constituents were melted several times to ensure homogeneity. The ingots were annealed in vacuum [torr] at for seven days and furnace cooled. Structural characterization was done by taking X-ray diffraction (XRD) patterns on sample powders employing Fe-K<sub>α</sub> radiation. Magnetization and Curie temperatures were determined using vibrating sample magnetometer (Model No PAR 155) up to an applied field of 12 kOe. For the determination of easy magnetization directions (EMDs), the sample powders were mixed with epoxy resin and were aligned in the presence of a magnetic field of 2.5 T applied perpendicular to the substrate. XRD patterns were taken on these compounds to determine the EMD.

**Results and discussions:**

X-ray diffraction patterns (Fig.1) show that all the compounds are for med in hexagonal (Th<sub>2</sub>Ni<sub>17</sub>) structure as shown by the representative planes in Figure 1. The lattice parameters calculated are given in Table I. There is a decrease in the unit cell volume with the substitution of Co for Fe in YGdFe<sub>17-x</sub>Si compounds which could be attributed to the small ionic radius of Co compared to that of Fe.

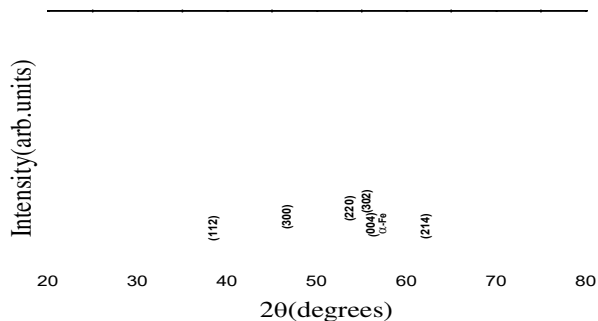


Fig.1. X-ray diffraction patterns of YGdFe<sub>17-x</sub>Co<sub>x</sub>Si (x=0, 1, 2 and 3) compounds.

Compound	a(Å)	c(Å)	V(Å <sup>3</sup> )	M <sub>s</sub> (emu/g)	T <sub>c</sub> ( K)
YGdFe <sub>17</sub> Si	8.61	8.4	622	130	410
YGdFe <sub>16</sub> CoSi	8.59	8.39	619	134	573
YGdFe <sub>15</sub> Co <sub>2</sub> Si	8.58	8.37	616	146	637
YGdFe <sub>14</sub> Co <sub>3</sub> Si	8.56	8.35	611	150	708

Table.1.lattice constants,saturation magnetization,and Curie temperature in  
(x=0,1,2And3)co  
mpounds.

The magnetization measurements done at 300K are shown in Figure.2. The saturation magnetization value were determined from Hondo plots and are given in Table I. The value of  $M_s$  is seen to increase from 130 emu/g for  $Y\text{GdFe}_{17}\text{Si}$  to 150 emu/g for  $Y\text{GdFe}_{14}\text{Co}_3\text{Si}$ . This increase in  $M_s$  could be due to the exchange between Co and Co and Fe and Co moments in addition to Fe-Fe exchange, based on the rigid band model.

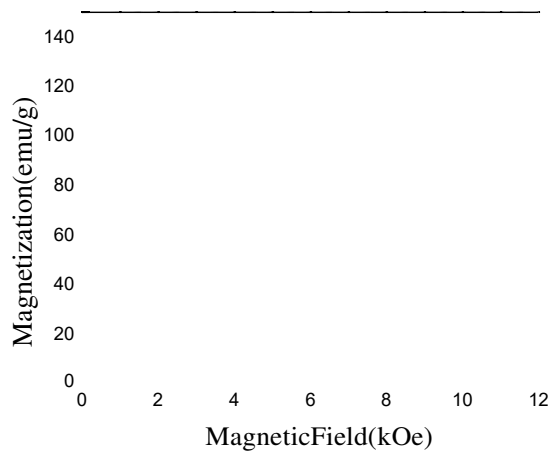


Figure.2.Magnetization curves of  $Y\text{GdFe}_{17-x}\text{Co}_x\text{Si}$  compounds measured at 80K.

The Curie temperature ( $T_c$ ) values were determined from M versus T measurements (Figure.3) in the presence of a magnetic field of 100 Oe. These values are found to increase to a large extent of about 288K from 410 K for  $Y\text{GdFe}_{17}\text{Si}$  to 708 K for  $Y\text{GdFe}_{14}\text{Co}_3\text{Si}$ . The strong ferromagnetic Fe-Co and Co-Co exchanges whose magnitudes are more than that of Fe-Fe exchange may cause the increment.

This increase in  $T_c$  values can also be explained on the basis of Friedel model [9], in which the interaction between two magnetic moments would be strong and ferromagnetic if the distance  $r_{ij}$  between them is smaller than the distance  $r_{ij}$  covered by the main peak of the Friedel oscillations, i.e., ( $r_{ij} < r_{ij}$ )

$\lambda/d > 1$ . In compounds containing 3d transition metals, it has been established that the magnetic coupling is governed mainly by the nearest-neighbor interactions and that is proportional to the lattice parameters.

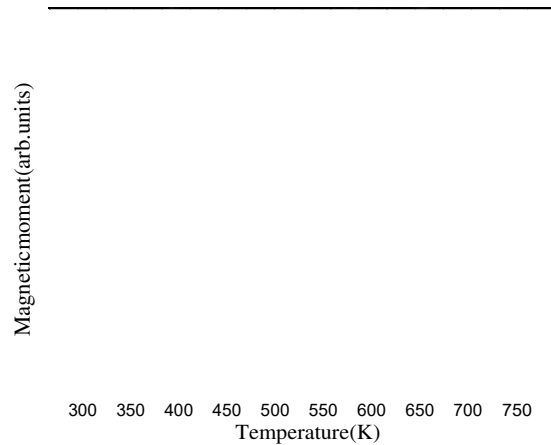


Figure.3. Temperature variation of magnetization of  $YGdFe_{17-x}Co_xSi$  compounds measured at 100 Oe.

Similarly, it has been shown that  $\lambda$  is inversely proportional to the  $d$  band Fermi wave vector,  $K_F$ . For the 3d band in the  $R_2Fe_{17}$  compounds,  $K_F$  is large. Substitution of Si decreases the holes in the 3d band and hence decreases  $K_F$ . Since the substitution of Si and Co brings about the reduction in the lattice parameters, there is hence an increase in the value of  $(\lambda/d)$  and  $T_C$ . Thus, the dual substitution of both Si and Co seems to improve the magnetic properties more effectively than the single substitution.

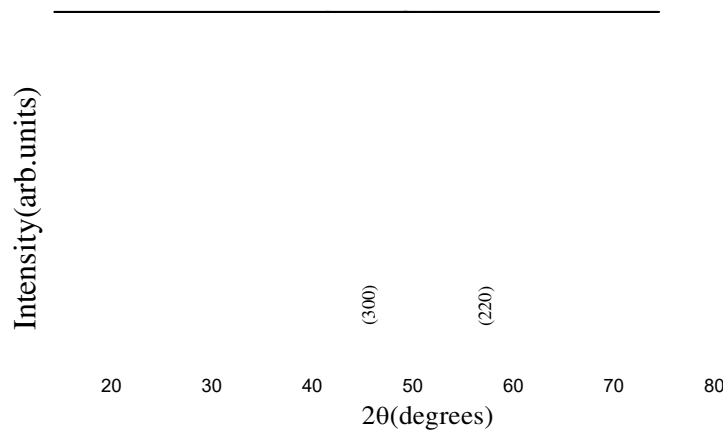


Figure.4. X-ray diffraction patterns of aligned  $YGdFe_{17-x}Co_xSi$  compounds.

The EMD of all the compounds were determined by taking the X-ray diffraction patterns on the aligned sample powders. Fig. 4 shows the XRD patterns for the aligned powders. It is observed that only two reflections (300) and (220) are seen. If the EMD is parallel to the ab-plane then only (hk0) reflections will be seen, and if the EMD is along the c-axis then only (00l) reflections will be intense. From the XRD patterns taken on the aligned samples shown in Fig. 4, the presence of planar anisotropy in these compounds is seen in all the compounds. Thus, the combined effect of Si and Co does not seem to cause any considerable change in the anisotropy.

### Summary:

The structural and magnetic properties of  $\text{YGdFe}_{17-x}\text{Co}_x\text{Si}$  were studied by X-ray diffraction and magnetization measurements. All the compounds are formed in hexagonal structure. Decrease in the cell volume is due to the smaller size of Co compared to that of Fe. There is a significant increase in the values of  $M_s$  and  $T_c$  in these compounds which are attributed to the additional Fe-Co, Co-Co exchanges along with Fe-Fe exchange, and also to the dual substitution of both Co and Si atoms.

### Acknowledgment:

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