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Formation, structure and hard magnetic properties of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds

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Abstract

The formation, crystal structure and magnetic properties of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ were studied. The substitution of Co for Fe helps to stabilize the rhombohedral $\text{Th}_2\text{Zn}_{17}$ -type structure (2:17) and eliminate α -Fe formation for carbon concentrations $y \leq 1.5$. From the mixing enthalpy calculations based on Miedema's semi-empirical model and Wigner–Seitz cell analysis, it is found that the role of Co in stabilizing the 2:17 structure is related to its strong preference for the 18h site, which has a maximum number of rare-earth near neighbours in the structure. With increasing Co content, x , the lattice constants decrease while the Curie temperature rapidly increases. The typical compound $\text{Sm}_2\text{Fe}_{12}\text{Co}_5\text{C}_{1.25}$ has a room temperature saturation magnetization $\mu_0 M_s = 1.36$ T, a magnetic anisotropy field $\mu_0 H_k = 7.1$ T, a Curie temperature $T_C = 874$ K and a possible maximum operating temperature T_m higher than 473 K.

1. Introduction

Nowadays, sintered and rapidly quenched NdFeB magnets are used widely due to their high energy products. However, the use of NdFeB magnets is generally limited to below 423 K due to their low Curie temperatures, T_C (about 585 K) [1, 2].

It is well known that $\text{Sm}_2\text{Fe}_{17}\text{C}_y$, $\text{Sm}_2\text{Fe}_{17}\text{N}_z$ and $\text{Sm}_2\text{Fe}_{17}\text{C}_y\text{N}_z$ intermetallic compounds with higher carbon or nitrogen concentrations have excellent intrinsic magnetic properties, i.e. a slightly lower magnetization $\mu_0 M_s$ (1.3–1.55 T), a higher magnetic anisotropy field $\mu_0 H_k$ at room temperature and a much higher T_C (~ 700 K) than $\text{Nd}_2\text{Fe}_{14}\text{B}$ [3–5]. Compounds of $\text{Sm}_2\text{Fe}_{17}\text{C}_y$ could be used as high-temperature magnets with a maximum operating temperature of 473 K. The carbon content, y , for 2:17 single-phase structural alloys prepared

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by conventional arc or induction melting methods is limited to less than 1.0 while higher carbon concentration is very important for gaining a high magnetic anisotropy field [4]. Alloys with higher carbon concentrations can be produced by the gas reaction method [5]. However, the high-temperature stability of the nitrides or carbides prepared by gas–solid reactions is rather poor; all the compounds disproportionate into α -Fe and SmC_x or SmN_x at temperatures above 973 K [6]. Shen *et al* [7] found that highly stable arc-melted $\text{Sm}_2\text{Fe}_{17}\text{C}_y$ compounds with $y = 1.5\text{--}3.0$ can be prepared by the partial substitution of Ga, Si, Al for Fe and can be good candidates for use as sintered hard magnets. However, the $\text{Sm}_2(\text{Fe}, \text{M})_{17}\text{C}_y$ compounds have relatively low saturation magnetization due to the introduction of non-magnetic M atoms.

Similar to the disproportionation behaviour of $\text{Sm}_2\text{Fe}_{17}\text{C}_y$ and $\text{Sm}_2\text{Fe}_{17}\text{N}_y$, the hydrides of $\text{Sm}_2\text{Fe}_{17}$ -based compounds will disproportionate into α -Fe and SmH_{2+x} at a temperature between 873 and 1173 K under a H_2 pressure of 0.1 MPa [8]. The addition of Ga, Co etc can increase the stability of $\text{Sm}_2(\text{Fe}, \text{Co}, \text{Ga})_{17}$ -type compounds in hydrogen [8–10]. So it may be possible to stabilize $\text{Sm}_2\text{Fe}_{17}\text{C}_y$ by the addition of some cobalt. In addition, the partial substitution of Co for Fe in $\text{Sm}_2\text{Fe}_{17}(\text{C}, \text{N})_y$ compounds will increase their T_C [11, 12]. In this work, we investigate the formation, structure and magnetic properties of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ alloys in order to identify a stable $\text{Sm}_2(\text{Fe}, \text{Co})_{17}\text{C}_y$ compound with good magnetic properties, which has potential to be a high-temperature permanent magnet with a superior performance to that of NdFeB-based alloys.

2. Experimental method

All alloys were prepared by tri-arc melting appropriate amounts of Fe (99.99%), Sm (99.9%), Co (99.9%) and Fe–C prealloy on a water-cooled copper boat under Ti-gettered prepurified Ar. The ingots were melted at least five times to ensure homogeneity. An excess of 15% Sm was added to the samples to partially compensate for the mass loss due to evaporation of Sm during the melting process. All ingots were wrapped in Ta foil and annealed in an evacuated quartz tube at 1353 K for one week.

X-ray diffraction (XRD) measurements on powder samples were performed on an automated Nicolet XRD with graphite monochromatized $\text{Cu K}\alpha$ radiation. The Curie temperatures, T_C , were measured by thermomagnetometry using a Perkin-Elmer thermogravimetric analyser (TGA-7) in a small magnetic field gradient. The error of T_C is less than 5 K. The thermomagnetic analysis method was also used to identify magnetic phases in the alloys. The magnetization, $M(H)$, was measured at room temperature using a vibrating sample magnetometer (VSM) with a maximum magnetic field of 1.8 T. The saturation magnetization, $\mu_0 M_s$, was determined by fitting the experimental $M(H)$ values plotted against $1/H$ using the law of approach to saturation. The aligned samples for anisotropy measurements were prepared by mixing the powders with epoxy resin and aligning in a magnetic field of 1.5 T. The easy axis direction was found from the XRD patterns of the magnetically aligned samples. The anisotropy field, $\mu_0 H_k$, was determined from magnetization curves along and perpendicular to the magnetically aligned direction.

3. Results and discussion

3.1. Formation and structure of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds

The phase constituents of the $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ alloys are presented in figure 1. In the figure, the curve represents the approximate phase boundary between the single-phase (2:17)

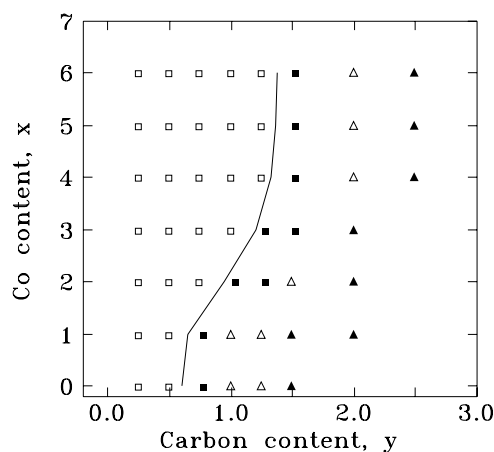


Figure 1. Phase constituents of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ alloys (empty squares—2:17 single phase; solid squares—2:17+ α -(Fe, Co), less than 5% by volume; empty triangles—2:17+ α -(Fe, Co), less than 25% by volume; solid triangles— α -(Fe, Co), more than 25% by volume; solid curve—approximate phase boundary between multi-phases and 2:17 single phase).

Table 1. Crystallographic data and Wigner–Seitz volume (‘WS vol.’) for $\text{Sm}_2\text{Fe}_{12}\text{Co}_5\text{C}_{0.75}$ (space group: $R\bar{3}m$; $a = 8.5301 \text{ \AA}$, $c = 12.4348 \text{ \AA}$).

Atom	Radius (\AA)	Site	Atomic position	Near neighbour	WS vol. (\AA^3)
Sm	1.81	6c	0, 0, 0.3428	1 Sm, 19 Fe	31.1
Fe	1.26	6c	0, 0, 0.0946	1 Sm, 13 Fe	12.3
Fe		9d	1/2, 0, 1/2	2 Sm, 10 Fe	11.2
Fe		18f	0.289, 0, 0	2 Sm, 11 Fe	11.6
Fe		18h	0.5020, -0.5020, 0.1540	3 Sm, 9 Fe	11.9
Co	1.25	18h	0.5020, -0.5020, 0.1540	3 Sm, 9 Fe	

composition region and that of the multi-phase (2:17, α -(Fe, Co), SmC etc). Alloys with $x \leq 2$ constitute the major phase, with rhombohedral, $\text{Th}_2\text{Zn}_{17}$ -type, structure (2:17). The formation of 2:17 structure becomes difficult and some α -(Fe, Co) forms when the carbon content rises to a critical value y_0 . A small amount of α -Fe (<5% by volume, determined from thermomagnetic measurements) is present in $\text{Sm}_2\text{Fe}_{17}\text{C}_{0.75}$ alloy prepared under our experimental conditions. However, the replacement of Fe by Co helps the formation of the 2:17 structure and the elimination of α -(Fe, Co), which is very useful for gaining a stable 2:17 phase structure in the alloys. With increasing Co content, x , the critical carbon concentration y_0 increases as well. When the Co content reaches $x = 5$, a single phase with the 2:17 structure is formed in the $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ alloys with $y = 1.25$. When the carbon content is $y = 1.5$, a small amount of α -Fe (<5% by volume) is present.

In the $\text{Sm}_2\text{Fe}_{17}\text{C}_y$ compounds, the Sm atoms occupy the 6c site, while the Fe atoms occupy four different sites, 6c, 9d, 18f and 18h. Neutron diffraction studies show that Co, Si, Al, Ga atoms exhibit a strong preference for substituting for Fe atoms in the 18h sites in $\text{Nd}_2\text{Fe}_{17}$ [13], which is isostructural to $\text{Sm}_2\text{Fe}_{17}\text{C}_y$. On the basis of the known atomic positions in the $\text{Nd}_2\text{Fe}_{17}$ compound [13], a Rietveld fit was used to refine the structure of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds by adjusting atomic positions, Debye–Waller factors, cell constants and the x-ray line profile parameters. The results are listed in table 1.

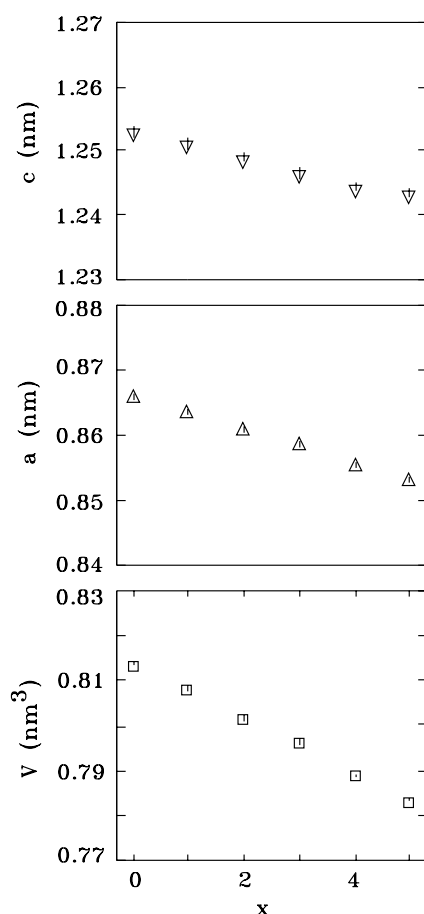


Figure 2. Lattice constants a , c and crystal cell volumes for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_{0.75}$ alloys.

While the atomic radius of Co (1.25 Å) is slightly smaller than that of Fe (1.26 Å) and the site volume of 18h is larger than that of the 18f and 9d sites (table 1), the Co atoms prefer to occupy the 18h site. The lattice parameters of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds decrease almost linearly with increasing Co content (figure 2). A similar result is found for $\text{Sm}_2(\text{Fe}, \text{Si})_{17}\text{C}_y$ [13–15]. The Si atoms enter the 18h site and cause the cell volume to decrease. Both Si and Co also cause the Curie temperature, T_C , to increase in the $\text{Sm}_2(\text{Fe}, \text{M})_{17}\text{C}_y$ compounds ($\text{M} = \text{Co}, \text{Si}$). However, the Co cannot stabilize 2:17 carbides with carbon content $y \geq 2.0$ as effectively as other stabilizing elements such as Si, Ga, Al.

The differences in stabilizing ability of Co, Si, Ga, Al in the 2:17 structure can be explained by calculating the mixing enthalpies on the basis of Miedema's model. In the model originally proposed by Miedema [16] and later improved [17, 18], the enthalpy of formation or enthalpy of mixing results from three factors. One is the difference between the work functions, Φ , of the pure metals. The second factor is the discontinuity of the electron density, n_{WS} , at the Wigner–Seitz cell boundary. The third factor, R , only occurs in alloys containing a transition metal and a polyvalent non-transition metal. The approximate formula for enthalpy of mixing can be expressed as follows [18]:

$$\Delta H^{\text{max}} = -P(\Delta\Phi)^2 + Q(\Delta n^{1/3})^2 - R \quad (1)$$

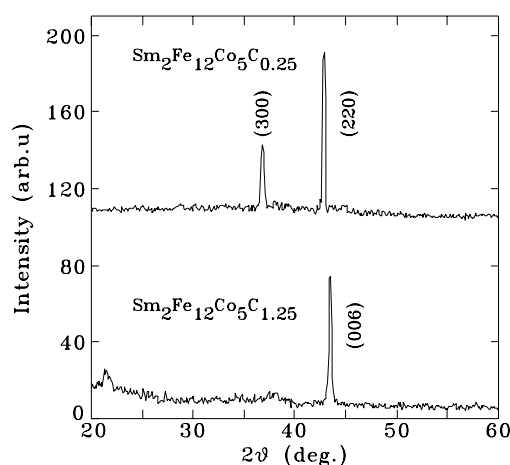


Figure 3. XRD patterns for magnetically aligned $\text{Sm}_2\text{Fe}_{12}\text{Co}_5\text{C}_y$ samples with $y = 0.25, 1.25$.

Table 2. Calculated mixing enthalpies for some binary alloys.

Alloy system	ΔH^{\max} (kJ mol ⁻¹)	Alloy system	ΔH^{\max} (kJ mol ⁻¹)
Sm-Fe	-9.16	Fe-Co	-0.36
Sm-Co	-22.1	Fe-Ga	-1.12
Sm-Ga	-26.6	Fe-Al	-7.05
Sm-Al	-30.6	Fe-Si	-5.51
Sm-Si	-38.2		

where P and Q/P are constant. In binary alloys, both elements are transition or non-transition elements; P is 14.1 and 10.7 respectively. If one of the atoms is a transition element and the other is a non-transition element, P is 12.4. The value of Q/P is 9.4.

Using equation (1) and the data in [18], we have calculated the enthalpy of mixing, ΔH^{\max} , between Sm and Fe, Co, Si, Ga, Al and that between Fe and Co, Si, Al. The results are listed in table 2. The ΔH^{\max} between Sm and Fe, Co, Si, Ga, Al are all negative and the values are much larger than those between Fe and Co, Si, Al, indicating that the Sm-(Fe, Co, Si, Ga, Al) bond is much stronger than the Fe-(Si, Co, Al, Ga) bond. The Wigner-Seitz cell analysis indicates that the 18h site has a maximum number of rare-earth near neighbours in the 2:17 compound (table 1). Thus the Co, Si, Ga, Al atoms enter the 18h site and stabilize the 2:17 structure. The values of ΔH^{\max} for Sm-Si, Sm-Ga, Sm-Al are about three to four times those for Sm-Fe, but ΔH^{\max} for Sm-Co is only double that for Sm-Fe. In addition, the ΔH^{\max} between Fe and Si, Ga or Al is larger than that between Fe and Co. This is the main reason that Co cannot stabilize 2:17 carbides as effectively as Si, Ga, Al etc.

3.2. Magnetic properties of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds

As mentioned above, a stable 2:17 single-phase structure is very important for preparing $\text{Sm}_2\text{Fe}_{17}\text{C}_y$ -based hard magnets. We concentrate in the discussion of magnetic properties on the alloys with a 2:17 single-phase structure (i.e. $y \leq 1.5$, as indicated in figure 1).

As a hard magnet, the compound should possess a strong easy c -axis magnetic anisotropy field. We use the XRD method to determine the easy axis direction of the compounds. As an example, the XRD patterns for magnetically aligned $\text{Sm}_2\text{Fe}_{12}\text{Co}_5\text{C}_y$ samples with

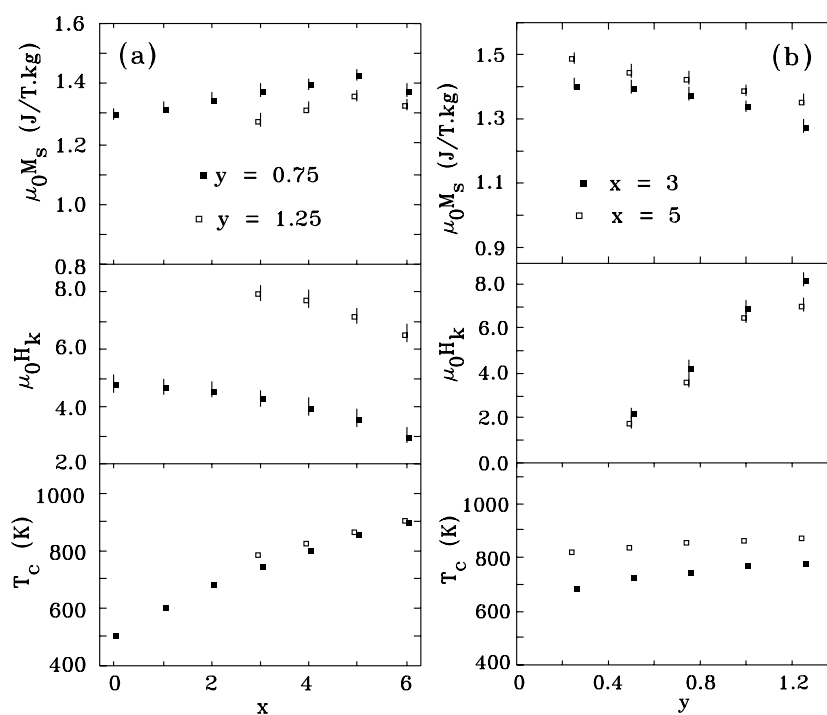


Figure 4. The room temperature saturation magnetization $\mu_0 M_s$, the Curie temperature T_C and the room temperature magnetic anisotropy field $\mu_0 H_k$ for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds as a function of Co content x (a) and of carbon content y (b) (The $\mu_0 H_k$ data for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_{0.25}$ alloys with $x = 3, 5$ are not shown because the alloys possess easy plane magnetic anisotropy.)

$y = 0.25, 1.25$ are shown in figure 3. For the $\text{Sm}_2\text{Fe}_{12}\text{Co}_5\text{C}_{1.25}$ compound, the intensity of the peak for the (006) crystal plane becomes very strong, and those for other peaks are very weak, which indicates a strong easy c -axis magnetocrystalline anisotropy. For $\text{Sm}_2\text{Fe}_{12}\text{Co}_5\text{C}_{0.25}$ alloy, the situation is reversed; the (220) and (300) peak intensities are very strong, which indicates an easy ab -plane anisotropy. The total XRD results for the aligned samples indicate that all $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ carbides with $x \leq 6$ and $y \geq 0.5$ have an easy c -axis magnetic anisotropy, while the compounds $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_{0.25}$ have an easy ab -plane anisotropy.

Figure 4(a) displays the room temperature saturation magnetization, $\mu_0 M_s$, the Curie temperature, T_C , and the room temperature magnetic anisotropy field, $\mu_0 H_k$, for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds with $y = 0.75, 1.25$ as a function of Co content. The Co content dependences of $\mu_0 M_s$, T_C and $\mu_0 H_k$ are similar for alloys with different carbon contents, $y = 0.75, 1.25$. The saturation magnetization increases with increasing Co content and reaches a maximum of about 1.36–1.40 T at $x = 5$, which can be understood on the basis of the Pauling–Slater curve. According to the Pauling–Slater curve, the maximum of $\mu_0 M_s$ in $\text{Fe}_{1-x}\text{Co}_x$ alloy appears at $x = 0.3$. T_C increases rapidly from about 500 K to 910 K with x increasing from 0 to 6. For the alloys with $x = 4–6$, T_C is about 830–910 K, which is much higher than that of the $\text{Nd}_2\text{Fe}_{14}\text{B}$ compound ($T_C = 585$ K). The anisotropy field decreases with increase in the Co content. $\mu_0 H_k$ for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_{1.25}$ is almost double that for the corresponding alloy with $y = 0.75$. This means that the $\mu_0 H_k$ strongly depends on the carbon content.

Figure 4(b) displays the room temperature saturation magnetization, $\mu_0 M_s$, the Curie temperature, T_C , and the room temperature magnetic anisotropy field, $\mu_0 H_k$, for

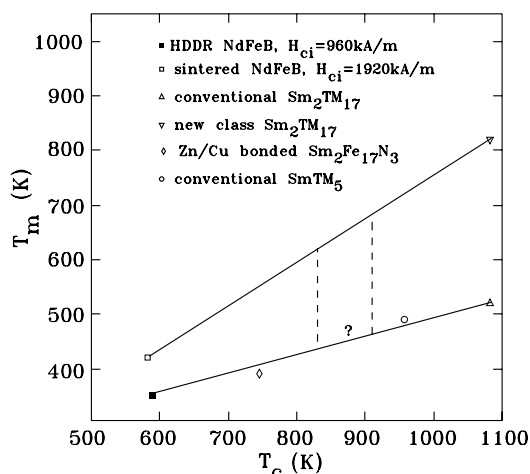


Figure 5. The relationship between the Curie temperature T_C and the maximum operating temperature T_m for hard rare-earth magnets (the data are cited from [19–21]; the dashed lines indicate T_C for $\text{Sm}_2\text{Fe}_{12}\text{Co}_x\text{C}_{1.25}$ with $x = 4$ (left) and $x = 6$ (right); the question mark stands for the T_C and a possible T_m of $\text{Sm}_2\text{Fe}_{12}\text{Co}_5\text{C}_{1.25}$ alloy).

$\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds with $x = 3, 5$ as a function of carbon content. The carbon content dependences of $\mu_0 M_s$, T_C and $\mu_0 H_k$ are similar for the two alloys. The saturation magnetization slightly decreases with increasing carbon content, y , due to the crystal cell expansion. The Curie temperature increases slowly with increase of the carbon content. Comparing with the results of figure 4(a), T_C is mainly determined by Co content for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ alloys with $y \geq 0.25$. However, $\mu_0 H_k$ increases rapidly with increasing carbon content. For $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ alloys with $y = 1.0$ – 1.25 , the room temperature $\mu_0 H_k$ is about 7–8 T, which is comparable with those of $\text{Nd}_2\text{Fe}_{14}\text{B}$ ($\mu_0 H_k = 6.7$ T) and $\text{Sm}_2\text{Co}_{17}$ ($\mu_0 H_k = 6.5$ T). In order to prepare a strong hard magnet, the carbon content should be higher than 1.0 for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ alloys. The large magnetocrystalline anisotropy originates from the effect of the interstitial carbon atoms for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ alloys with $x \leq 6.0$, because the alloys of $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x$ show easy plane anisotropy for $x \leq 8.5$. A typical compound of $\text{Sm}_2\text{Fe}_{12}\text{Co}_5\text{C}_{1.25}$ has a $\mu_0 M_s$ of about 1.36 T, a T_C of 874 K, a room temperature $\mu_0 H_k$ of about 7.1 T; these values are better than or comparable to those for the $\text{Nd}_2\text{Fe}_{14}\text{B}$ compound.

Figure 5 shows the relationship between the Curie temperature T_C and the maximum operating temperature T_m for RE–TM (TM = Fe, Co) magnets, which is constructed according to the data on $\text{Nd}_2\text{Fe}_{14}\text{B}$ -, $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ -, SmCo_5 - and $\text{Sm}_2\text{Co}_{17}$ -based magnets [19–21]. T_m increases nearly linearly with increasing T_C . However, some magnets maybe have very different values of T_m due to their different microstructures and coercivities although they are composed of the same magnetic compound. For example, $\text{Sm}_2\text{Co}_{17}$ -based conventional and new-type high-temperature $\text{Sm}_2\text{TM}_{17}$ (TM = Co, Fe, Cu, Zr) magnets have T_m equal to about 523 and 823 K respectively [19]. As mentioned above, the compound $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_{1.0-1.25}$ possesses a room temperature $\mu_0 H_k$ of 7–8 T, comparable to those of $\text{Nd}_2\text{Fe}_{14}\text{B}$ and $\text{Sm}_2\text{Co}_{17}$. In addition, the temperature dependence curves of $\mu_0 H_k$ for $\text{Nd}_2\text{Fe}_{14}\text{B}$ and $\text{Sm}_2\text{Fe}_{17}\text{C}_y$ have similar slopes [5]. It is reasonable to assume that the relation between T_m and T_C for $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_{1.0-1.25}$ conforms to the relation shown in figure 5. The potential magnets based on $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_{1.0-1.25}$ with $x = 4$ – 6 have T_C between 830 and 910 K and should possess a T_m higher than 473 K, which is between those of NdFeB and $\text{Sm}_2\text{TM}_{17}$ (figure 5).

The potential magnet could be operated in environments which need a T_m larger than that of NdFeB, but less than that of the $\text{Sm}_2\text{TM}_{17}$ magnet. In particular, the potential magnet could replace the SmCo magnet in some fields and reduce the cost.

4. Conclusions

In summary, the compounds $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ with $x = 4-6$ and $y = 1.0-1.25$ possess a room temperature $\mu_0 M_s$ of about 1.35–1.40 T, a T_C of about 830–910 K, a room temperature $\mu_0 H_k$ of about 7 to 8 T and a possible maximum operating temperature T_m higher than 473 K. These good intrinsic magnetic properties strongly suggest that this compound could be used as a hard magnetic material with a much higher operating temperature than NdFeB.

The $\text{Sm}_2\text{Fe}_{17-x}\text{Co}_x\text{C}_y$ compounds with rhombohedral $\text{Th}_2\text{Zn}_{17}$ -type single-phase structure can be prepared using conventional arc-melting technology while the carbon concentration, y , is kept moderate ($y = 1.0-1.25$), because the substitution of Co for Fe helps to stabilize the 2:17 structure in the alloys. This allows a conventional sintering process to be used for manufacturing practical hard magnets based on the above compounds with a higher operating temperatures than for sintered NdFeB magnets but at a lower cost than Sm–Co magnets.

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