

Overview of Recent Progress in Sm-Co Based Magnets

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Abstract: Superior magnetic properties of Sm-Co based magnets, especially their excellent high temperature stability and low temperature coefficient, have led to an exciting variety of applications. Despite the commercial success of sintered Nd-Fe-B magnets, Sm-Co magnets continue to play a dominant role in some critical applications, such as traveling wave tubes (TWT) for space exploration and satellite communication; inertial devices for accelerometers and gyroscopes; power tools for medical applications; and permanent magnet motors and generators for aircraft engines. We will review the recent development in ultra-high temperature magnets and zero RTC (reversible temperature coefficient) magnets.

Key words: SmCo magnets, high temperature magnets, zero reversible temperature coefficient (RTC)

1. Introduction

Rare earth permanent magnets have attracted significant attention in a variety of applications due to their high magnetic performance and availability. There are three families of rare earth magnets commercially available, namely SmCo_5 , $\text{Sm}_2\text{Co}_{17}$ and $\text{Nd}_2\text{Fe}_{14}\text{B}$ based magnets, which were developed in the 1960s, 1970s and 1980s, respectively.

The development of SmCo_5 based magnets is a result of the research on the anisotropy of YCo_5 in mid-1960s^[1]. Liquid phase sintering technique proved to be the key for the development of fully dense and stable SmCo_5 magnets^[2], which became the first generation of rare earth magnets. SmCo_5 magnets, with a maximum energy product of up to 18 MGOe and high intrinsic coercivity (iH_c) to resist thermal and field demagnetization, are still widely used in many applications.

$\text{Sm}_2\text{Co}_{17}$ -based magnets were an outgrowth of the investigation of $\text{R}_2(\text{Co}, \text{Fe})_{17}$ alloys in the 1970s in an effort to increase the maximum energy product, $(\text{BH})_{\text{max}}$. However, the development of high intrinsic coercivity proved to be difficult even though the quasi-binary $\text{R}_2(\text{Co}_{1-x}\text{Fe}_x)_{17}$ intermetallics with reasonable anisotropy exists in most of the light rare earth systems. In the mid 1970s, research on Cu- and Zr-added $\text{Sm}(\text{Co}, \text{Fe}, \text{Cu}, \text{Zr})_z$ alloys resulted in a $(\text{BH})_{\text{max}}$ as high as 26 to 30 MGOe^[3], which led to the commercial success of the 2nd generation of rare earth magnets.

Extensive research was conducted in the late 1970s

and early 1980s to improve the magnetic performance of SmCo magnets by replacing Sm with other light rare earth elements, such as Nd and Pr, and replacing Co with Fe, which led to the development of the third generation, $\text{Nd}_2\text{Fe}_{14}\text{B}$ -based, rare earth magnets with superior room temperature magnetic performance^[4]. Although the intrinsic coercivity and Curie temperature of $\text{Nd}_2\text{Fe}_{14}\text{B}$ -based magnets are inferior to those of Sm-Co based magnets, there is a great improvement in room temperature residual induction.

The discovery of $\text{Nd}_2\text{Fe}_{14}\text{B}$ -based magnets sparked more interest in searching for other novel magnet materials in the late 1980s and 1990s. It was discovered that ThMn_{12} -type intermetallics with a general formula of $\text{RFe}_{12-x}\text{M}_x$ ($\text{M}=\text{Ti}, \text{V}, \text{Cr}, \text{Mo}$ and Si) have attractive properties although there are still some difficulties in producing consistently superior magnets for commercial applications. There were also extensive studies of $\text{Sm}_2\text{Fe}_{17}\text{N}_x$ in the 1990s but the properties deteriorate when heated to temperatures beyond 600°C, which prevent it from becoming a sintered fully dense magnet. Nanocomposite rare earth magnets also attracted significant attention in the 1990s in order to take advantage of the high saturation magnetization of soft magnetic phases and the high anisotropy of the hard magnetic phases^[5-9]. Currently, there is still an on-going effort worldwide for the development of high performance nanocomposite magnets. In this paper, we will review the recent progress on SmCo based permanent magnets with ultra-high temperature stability and small reversible

temperature coefficient of B_r .

2. Experimental Procedures

A series of samples with a nominal composition of $(Sm_{1-x}Gd_x)(Co_{0.71}Fe_{0.18}Cu_{0.08}Zr_{0.027})_7$ ($x=0-0.55$) were made with a conventional powder metallurgy process. Axially or iso-statically pressed green compacts were sintered at 1195 to 1225°C, followed by a homogenization treatment between 1160 to 1185°C. All samples were then aged between 820 to 890°C for 10 hours followed by a slow cooling at 1°C/min. to 400°C. The samples were then machined into cylinders measuring 0.393 inches in diameter and 0.393 inches in thickness. Magnetic properties were tested using a KJS Hysteresigraph and a Vibrating Sample Magnetometer (VSM) at various temperatures from -100 to 400°C. The reversible temperature coefficient of B_r , α , for each composition was calculated in the temperature range between -50°C and 150°C for the convenience of comparison.

3. Results and Discussion

Prior to 1997, the best high temperature magnets available were limited to applications where temperatures did not exceed 300°C. The US Air Force funded development work to increase high temperature performance of magnets under MURI and STTR programs in response to the More Electric Aircraft (MEA) Initiative, which led to significant progress in improving the high temperature properties of sintered rare earth-Co based magnets^[10-16]. As a result, EEC developed and patented a series of ultra-high temperature magnets with maximum operating temperatures up to 550°C. Table 1 shows the typical magnetic properties of ultra-high temperature magnets.

Table 1. Typical magnetic properties and maximum operating temperature (T_M) of high temperature magnets

Material	B_r	H_c	H_{ci}	$(BH)_{max}$	T_M (°C)
SmCo T550	8.5	7.9	>25	16	550
SmCo T500	9.2	8.9	>25	20	500
SmCo T450	9.6	9.1	>25	22	450
SmCo T400	10.1	9.5	>25	24	400

Some applications, such as inertial devices and traveling wave tubes, require small reversible temperature coefficient (RTC) of B_r , α , as well as high temperature stability. The currently best available temperature compensated RE(Co,Fe,Cu,Zr)_z magnets (EEC-2:17TC-16) with a maximum operating temperature (T_M) of 300°C have the following nominal magnetic properties: $B_r = 8.3$ kG, $H_{ci} > 25$ kOe, $H_c = 7.8$ kOe, $(BH)_{max} = 16$ MGOe, α (-50°C to 150°C) = -0.001%/°C.

The need for temperature compensated magnets with $T_M > 300$ °C prompted efforts to develop a temperature-compensated, high-temperature magnet series with a small RTC of B_r . In the past, various heavy rare earth elements such as Tb, Dy, Ho, Er, and Gd have been evaluated for partial Sm substitution to reduce the reversible temperature coefficient of B_r . In this work, the effects of Gd-substitution were studied on magnetic properties and α (RTC of B_r) of the TC400 high temperature Sm₂TM₁₇ magnet series with $T_M = 400$ °C.

Table 2. Typical magnetic properties, maximum operating temperature (T_M), and RTC of B_r (α) of $(Sm_{1-x}Gd_x)(Co_{0.71}Fe_{0.18}Cu_{0.08}Zr_{0.027})_7$ magnets

Magnet ID	x	Temp. °C	B_r (kG)	BH_{max} (MGOe)	α %/°C	T_M °C
T400-22	0.00	25	9.6	21.9	-0.037	400
		150	9.1	19.6		
		400	8.1	14.7		
TC400-20	0.09	25	9.1	19.9	-0.034	400
		150	8.7	18.0		
		400	7.8	13.6		
TC400-18	0.19	25	8.7	18.0	-0.030	400
		150	8.4	16.5		
		400	7.6	12.7		
TC400-16	0.28	25	8.3	16.5	-0.020	400
		150	8.1	15.3		
		400	7.4	12.1		
TC400-14	0.37	25	7.9	14.7	-0.015	400
		150	7.7	14.0		
		400	7.0	11.0		
TC400-13	0.46	25	7.4	13.0	-0.007	400
		150	7.3	12.5		
		400	6.8	10.3		
TC400-11	0.55	25	6.9	11.5	+0.002	400
		150	7.0	11.3		
		400	6.5	9.5		

The magnetic properties at 25°C, 150°C, and 400°C

for some TC400 magnets with a composition of $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_{0.71}\text{Fe}_{0.18}\text{Cu}_{0.08}\text{Zr}_{0.027})_7$ ($0 \leq x \leq 0.55$) are shown in Table 2. The corresponding reversible temperature coefficients of B_r (α) are also listed in Table 2 for comparison.

The reversible temperature coefficient of B_r (α) is a quantity that represents the rate of change in residual induction, B_r , in a specified temperature range. It is calculated using the following formula: $\alpha = \Delta B_r / B_r / \Delta T$, where $\Delta T = (T_2 - T_1)$, $B_r = B_r(T_1)$, $\Delta B_r = B_r(T_2) - B_r(T_1)$, the residual induction at temperature T_1 and T_2 is $B_r(T_1)$ and $B_r(T_2)$, respectively. For the convenience of comparison with conventional permanent magnets, the RTC of B_r is calculated in the temperature range between -50°C to $+150^\circ\text{C}$.

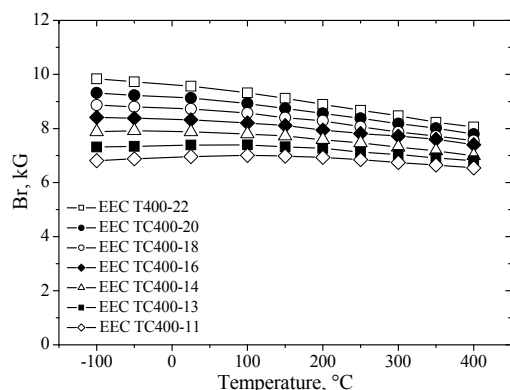


Figure 1. Effect of temperature on the residual induction (B_r) of the $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_y\text{Fe}_u\text{Cu}_v\text{Zr}_w)_7$ magnets

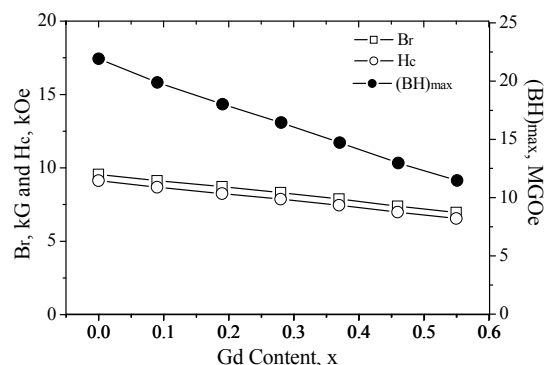


Figure 2. Effect of Gd content (x) on the magnetic properties of the $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_y\text{Fe}_u\text{Cu}_v\text{Zr}_w)_7$ high temperature magnets

As shown in Table 1, the magnetic properties

decrease with the increase of the Gd content x , which is expected, because $\text{Gd}_2\text{Co}_{17}$ has a lower saturation magnetization compared to $\text{Sm}_2\text{Co}_{17}$ phase. Room temperature residual induction, B_r , decreases from 9.6 kG to 6.9 kG when the Gd content, x , changes from 0 to 0.55 in the $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_y\text{Fe}_u\text{Cu}_v\text{Zr}_w)_7$ magnets.

Figure 1 shows the residual induction B_r as a function of temperature for various compositions. It is interesting to note that the residual induction, B_r , of TC400-11 magnets changes from 6.88 kG to 6.54 kG when the temperature changes from -100 to $+400^\circ\text{C}$. Such a small variation in B_r within a wide temperature window could lead to new applications.

The room temperature magnetic properties of various TC400 magnets are shown in Figure 2. The residual induction, B_r , the coercivity, H_c , and the maximum energy product, $(\text{BH})_{\text{max}}$, decrease with the increase of the Gd content, x , in $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_y\text{Fe}_u\text{Cu}_v\text{Zr}_w)_7$ magnets. Depending upon the requirements on the reversible temperature coefficient of B_r , the TC400 high temperature magnet series can offer a range of $(\text{BH})_{\text{max}}$ from 11.5 MGOe to 22 MGOe. Figures 3, 4, and 5 are typical demagnetization curves of the T400-22, TC400-16, and TC400-11 magnets, respectively.

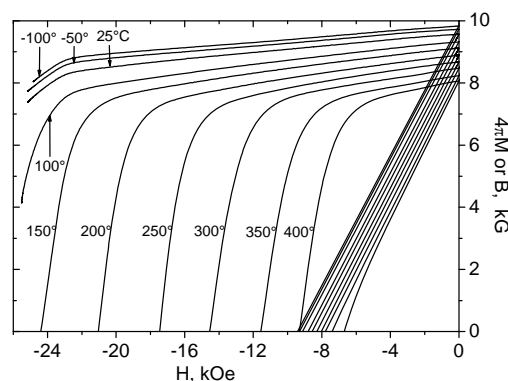


Figure 3. Typical demagnetization curves of T400-22 high temperature magnets

The TC400 magnet series are perfect high temperature magnets with a T_M of 400°C because of their square intrinsic demagnetization curves and the straight-line extrinsic demagnetization curves over the entire temperature range up to 400°C . The addition of Gd does not have an adverse effect on the

demagnetization curve shape, while it has the benefit of temperature compensation.

As shown in Figure 6, the reversible temperature coefficient of B_r is almost linearly proportional to the Gd content, x , in the $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_y\text{Fe}_u\text{Cu}_v\text{Zr}_w)_7$ magnets. This would allow us to calculate the values of α for a predetermined Sm/Gd ratio in the TC400 magnet series.

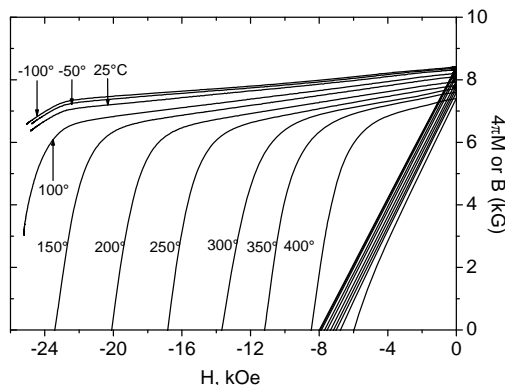


Figure 4. Typical demagnetization curves of TC400-16 high temperature magnets

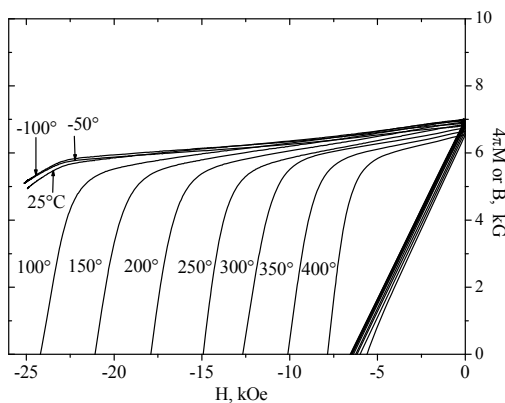


Figure 5. Typical demagnetization curves of TC400-11 high temperature magnets

The studies on the effects of partial Gd substitution on the magnetic properties and RTC of B_r resulted in a new TC400 series of temperature-compensated high-temperature permanent magnets. When the Gd content, x , is about 0.55 in the $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_y\text{Fe}_u\text{Cu}_v\text{Zr}_w)_7$ magnets, the reversible temperature coefficient (RTC) of B_r (α) is almost zero.

The residual induction of TC400-11 magnets

changes by only 0.34 kG when the temperature changes from -100 to $+400^\circ\text{C}$. This represents a reversible temperature coefficient of B_r of only $-0.01\%/^\circ\text{C}$ within the temperature range from -100°C to $+400^\circ\text{C}$. For comparison, its RTC of B_r is $+0.002\%/^\circ\text{C}$ within the temperature range from -50°C to $+150^\circ\text{C}$. The remarkable high temperature stability over such a broad temperature window could lead to new applications.

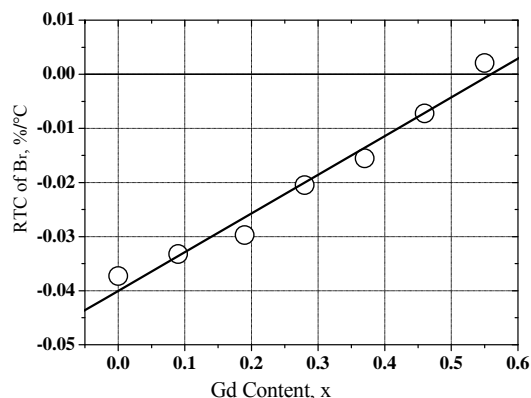


Figure 6. Effect of Gd content (x) on the RTC of B_r (α) for the TC400 magnet series with a nominal composition of $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_y\text{Fe}_u\text{Cu}_v\text{Zr}_w)_7$

All TC400 series magnets have straight extrinsic demagnetization curves up to 400°C regardless of the Gd content, which is consistent with the belief that the maximum operating temperature depends on the Co content rather than Gd content. Because the Co content was kept constant, all TC400 magnets have the same maximum operating temperature.

5. Summary

The maximum energy product, $(\text{BH})_{\text{max}}$, is about 24, 20, and 16 MGOe for ultra-high temperature SmCo magnets with maximum operating temperatures of 400, 500 and 550°C , respectively. Further development work shows that the reversible temperature coefficient of B_r can be reduced to almost zero while maintaining the maximum operating temperature of 400°C .

A TC400 series of temperature-compensated

high-temperature permanent magnets was developed by replacing partially Sm with Gd in $(\text{Sm}_{1-x}\text{Gd}_x)(\text{Co}_y\text{Fe}_u\text{Cu}_v\text{Zr}_w)_7$ magnets. When the Gd content x is about 0.55, the reversible temperature coefficient (RTC) of B_r (α) is almost zero. The TC400 magnet series has a maximum operating temperature (T_M) of 400°C, but with a significantly reduced RTC of B_r . The design of the TC400 magnet series was based on the current EEC-T400 high temperature magnets. With the increase of Gd content x from 0 to 0.55, the RTC of B_r , α , is reduced from -0.035%/°C to +0.002%/°C without a compromise in the high temperature capability of the magnets, while the room temperature residual induction, B_r , decreased from approximately 9.5 kG to 6.5 kG. All TC400 series temperature-compensated high temperature magnets have straight-line extrinsic demagnetization curves up to 400°C.

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