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Effect of Iron Substitution on the High-temperature Properties of $\text{Sm}(\text{Co,Cu,Ti})_z$ Permanent Magnets

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ABSTRACT

Recently, Ti-substituted Sm-Co permanent magnets have attracted renewed attention due to their interesting high-temperature coercivity. Our presentation deals with the effect of iron substitutions on the magnetic properties of the materials. X-ray diffraction shows that the investigated $\text{Sm}(\text{Co,Fe,Cu,Ti})_z$ materials ($z = 7.0 - 7.6$) are two-phase magnets, consisting of 1:5 and 2:17 regions. The iron content affects both the coercivity and the magnetization. Depending on composition and heat treatment, some samples show a positive temperature coefficient of the coercivity in the temperature range from 22 °C to 550 °C. Moderate amounts of iron enhance the room-temperature coercivity. For example, the room-temperature coercivity of $\text{Sm}(\text{Co}_{6.0}\text{Fe}_{0.4}\text{Cu}_{0.6}\text{Ti}_{0.3})$ is 9.6 kOe, as compared to 7.6 kOe for $\text{Sm}(\text{Co}_{6.4}\text{Cu}_{0.6}\text{Ti}_{0.3})$. At high temperatures, the addition of Fe has a deteriorating effect on the coercivity, which is as high as 10.0 kOe at 500 °C for $\text{Sm}(\text{Co}_{6.4}\text{Cu}_{0.6}\text{Ti}_{0.3})$. The room-temperature magnetization increases on iron substitution, from 73 emu/g for $\text{Sm}(\text{Co}_{6.4}\text{Cu}_{0.6}\text{Ti}_{0.3})$ to 78 emu/g for $\text{Sm}(\text{Co}_{6.0}\text{Fe}_{0.4}\text{Cu}_{0.6}\text{Ti}_{0.3})$. The observed temperature dependence is ascribed to the preferential dumbbell-site occupancy of the Fe atoms.

INTRODUCTION

The demand for permanent magnets with high-temperature applications above 450 °C has attracted much attention in recent years. Usually, the best room-temperature permanent magnets are made by Nd-Fe-B since it has a relatively high saturation magnetization and moderate intrinsic coercivity. These two properties lead to a high energy product $(\text{BH})_{\text{max}}$, which is a key figure of merit for a hard magnet. However Nd-Fe-B has large temperature coefficients of H_c and B_r . Therefore, $(\text{BH})_{\text{max}}$ drops below 10 MGOe when $T > 200$ °C, and even more for higher temperatures, which makes it unsuitable for high-temperature applications. Another type of commercially available permanent magnets is 2:17-type Sm-Co magnets. The complicated heat-treatment of $\text{Sm}(\text{Co, Fe, Cu, Zr})_z$ leads to a cellular microstructure that is Fe-containing SmCo-2:17 phases surrounded by Cu-rich SmCo_5 grain boundaries. Zr plays a key role in the formation of this type of structure by helping the precipitation of the Cu-rich phase. Although the maximum energy product of $\text{Sm}_2\text{Co}_{17}$ is not as high as that of the Nd-Fe-B at room temperature, the low temperature coefficients of H_c and B_r make the energy product larger at elevated temperature, which makes the 2:17 type Sm-Co based magnets a good choice for high-temperature applications [1, 2].

Our previous studies have been focused on SmCo-based alloys with Ti-substitution and their possible use as high temperature permanent magnets [3, 4]. An abnormal temperature dependence of coercivity has been found and an explanation for that has been given. An H_c of

12.3 kOe at 500 °C has been obtained. However, the relatively low magnetization limits the attainable energy product of these alloys. This paper reports on the Fe effects of substitution for Co on both coercivity and magnetization.

EXPERIMENTAL METHODS

$\text{Sm}(\text{Co}, \text{Fe}, \text{Cu}, \text{Ti})_z$ ($Z = 7.0-7.6$) are prepared by arc melting the starting element materials under flowing argon. The composition is represented in the form of $\text{Sm}(\text{Co}_{z-x-y}\text{Fe}_x\text{Cu}_y\text{Ti}_v)$ where $x = 0 - 1.0$, $y = 0.4-0.9$, and $v = 0.25, 0.3$. The as-melted samples are heat treated following a procedure similar to that of the $\text{Sm}_2\text{TM}_{17}$ magnets: homogenization at 1165 °C for 3 hours, annealing at 825 °C for 8 hours, slow cooling under a rate of 1 °C/min till 550 °C, and holding there for another 8 hours. Aligned samples are made by mixing ground powders with high-temperature cement in a 1.4 T field until solidification. X-ray diffraction is used for the determination of the crystal structure. SQUID magnetometer and high-temperature VSM are used to obtain magnetization data.

RESULTS AND DISCUSSION

The as-melted samples show the disordered 1:5 structure and after annealing the cellular-like microstructure forms, which contains a 5-10 nm thick grain-boundary Cu-rich 1:5 phase and a main phase with the 2:17 structure and with an average grain size of around 70 nm. This was shown in our previous work [3].

Table I shows the room temperature M_s , M_r , and H_c values for different Cu and Fe contents (TM/Sm ratio $z = 7.6$). All data show that additional Fe helps to increase M_s . For low Cu content (Cu = 0.6), more Fe decreases H_c . For high Cu content value, large coercivities are obtained and some samples are not saturated in a field of 55 kOe.

$Z=7.6$		M_s (emu/g)			M_r (emu/g)			H_c (kOe)		
Fe	Cu	0.6	0.7	0.8	0.6	0.7	0.8	0.6	0.7	0.8
0		70	66	63	48	41	43	10.6	7	30
0.4		78	77	68	60	64	45	8.7	18.2	23
0.7		82	N/s	N/s	39	N/s	N/s	4.5	N/s	N/s
1.0		89			58			3.8		

Table I. Room temperature M_s , M_r and H_c of $\text{SmCo}_{7.3-x-y}\text{Fe}_x\text{Cu}_y\text{Ti}_{0.3}$. (N/s means not saturated at 55 kOe)

For some particular compositions, a certain amount of Fe can increase both H_c and M_s . Figure 1 shows the temperature dependence of coercivity for the series of $\text{SmCo}_{6.4-x}\text{Fe}_x\text{Cu}_{0.6}\text{Ti}_{0.3}$. For $x \leq 0.7$, the samples show a positive temperature coefficient of coercivity (TCC). We see that on Fe substitution for Co the room temperature coercivity reaches a maximum at a certain Fe

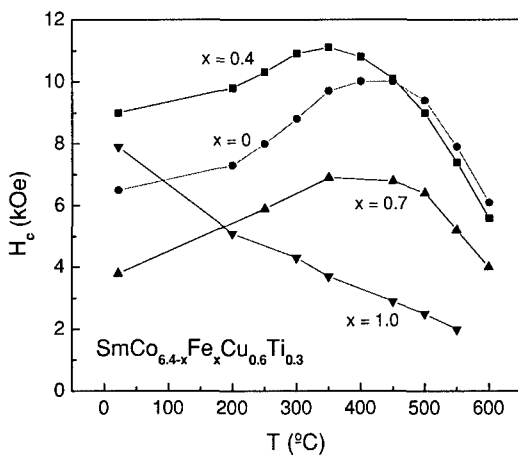


Figure 1. Temperature dependence of coercivity of $\text{SmCo}_{6.4-x}\text{Fe}_x\text{Cu}_{0.6}\text{Ti}_{0.3}$.

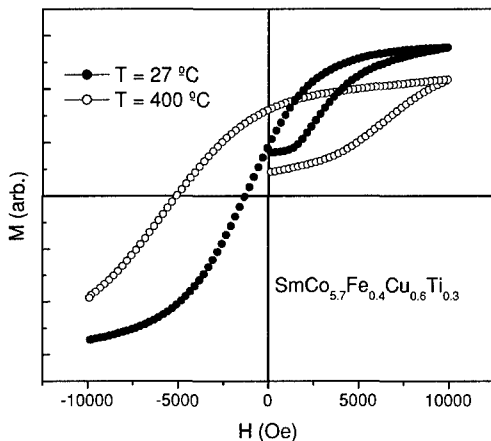


Figure 2. Typical loops for a sample with positive TCC at room temperature and 400 °C.

amount, then decreases while the TCC remains positive. On further addition of Fe, the TCC becomes negative and also the coercivity drops to a relatively low level. The positive TCC has been explained by a model in terms of domain-wall-pinning mechanism in a two-phase magnet [4].

As in the case of conventional SmCo 2:17 magnets, Fe substitutes for Co and most likely enters the main 2:17 phase in the cellular structure, and also stabilizes the 2:17 phase at the

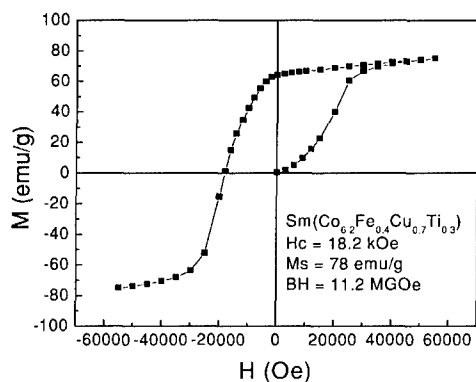


Figure 3(a). Hysteresis loop of $\text{SmCo}_{6.2}\text{Fe}_{0.4}\text{Cu}_{0.7}\text{Ti}_{0.3}$ at room-temperature.

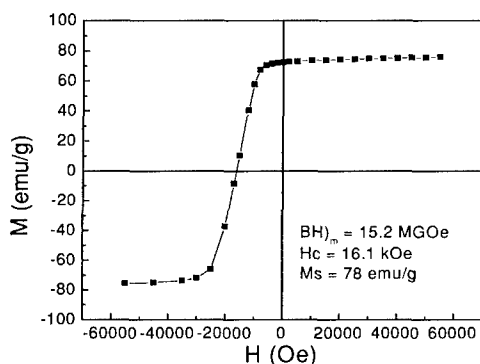


Figure 3(b). Hysteresis loop of aligned $\text{SmCo}_{6.2}\text{Fe}_{0.4}\text{Cu}_{0.7}\text{Ti}_{0.3}$ sample at room-temperature.

presence of Cu [5]. The increase of the room-temperature coercivity due to Fe is explained by the preferential dumbbell-site occupation of Fe in the 2:17 structure [6]. This enhances the anisotropy of the 2:17 phase and results in a higher coercivity as compared to the Fe-free sample. Figure 2 shows typical loops for a sample with positive TCC at room temperature and 400 °C. Even though at 400 °C the sample is not saturated, it is obvious that the coercivity increases with increasing temperature. This is due to the different temperature dependence of the two phases (1:5 and 2:17). A detailed explanation of this mechanism has been given elsewhere [4].

Figure 3(a) shows the room-temperature loop of a sample, whereas 3(b) shows the same sample after alignment. The energy product has been improved from 11.2 MGOe to 15.2 MGOe. The demagnetization curve of the $\text{SmCo}_{6.2}\text{Fe}_{0.4}\text{Cu}_{0.7}\text{Ti}_{0.3}$ bulk sample at room temperature and 500 °C is shown in Fig. 4. A coercivity of 7.3 kOe and an energy product of 5.3 MGOe is obtained. Noticing that the grains in the sample are randomly oriented, a higher energy product is anticipated in aligned or sintered magnets. Because we do not have a high temperature

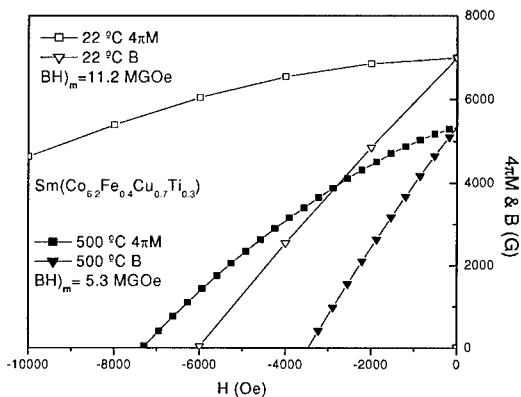


Figure 4. Demagnetization curves for $\text{SmCo}_{6.2}\text{Fe}_{0.4}\text{Cu}_{0.7}\text{Ti}_{0.3}$ bulk sample.

epoxy that can withstand 500 °C, no results for aligned samples have been obtained at this temperature until now. Further investigations on sample alignment are still under way, but the good property shown in bulk sample makes the material promising for high-temperature application as a permanent magnet.

Figure 5 shows the change of coercivity at room temperature and 500 °C. We can see that for $\text{Fe} = 0.7$, the high-temperature coercivity remains around 8.5 kOe. This makes it possible to increase remanence by introducing certain amount of Fe without losing too much coercivity. That will lead to obtain an energy product value around 10 MGOe at 500 °C.

Figure 6 shows the demagnetization curves for $\text{SmCo}_{6.0}\text{Fe}_{0.4}\text{Cu}_{0.6}\text{Ti}_{0.3}$ at different temperatures. The increase in H_c is obvious and an H_c of 9 kOe is obtained at 500 °C.

CONCLUSIONS

Fe substitution for Co enhances the magnetization of Sm-Co-Cu-Ti and, for some compositions, the coercivity. The latter is due to the preference of the Fe occupation of dumbbell site in the 2:17 phase. The room-temperature coercivity of $\text{Sm}(\text{Co}_{6.0}\text{Fe}_{0.4}\text{Cu}_{0.6}\text{Ti}_{0.3})$ is 9.6 kOe, as compared to 7.6 kOe for $\text{Sm}(\text{Co}_{6.4}\text{Cu}_{0.6}\text{Ti}_{0.3})$. However, larger Fe amounts ($\text{Fe} \geq 1.0$) will decrease the coercivity at both room temperature and elevated temperature. An H_c of 9.0 kOe is obtained at 500 °C. The substitution of Fe for Co helps increase the magnetization at all the temperatures, which leads to a higher energy product than that of Fe-free samples. A $(\text{BH})_{\text{max}}$ of 11.2 MGOe and 5.3 MGOe in a $\text{SmCo}_{6.2}\text{Fe}_{0.4}\text{Cu}_{0.7}\text{Ti}_{0.3}$ bulk sample is obtained at room temperature and 500 °C, respectively. A large increase of $(\text{BH})_{\text{max}}$ is anticipated in aligned samples. These properties make this material a promising candidate for high-temperature applications.

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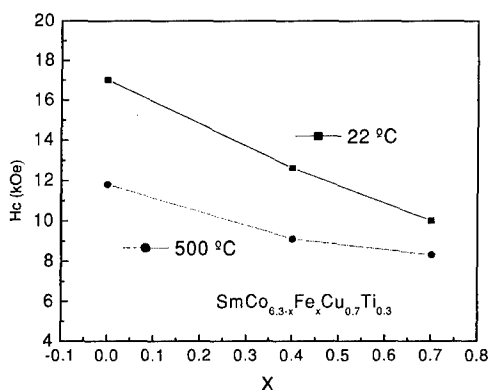


Figure 5. Coercivities of $\text{SmCo}_{6.3-x}\text{Fe}_x\text{Cu}_{0.7}\text{Ti}_{0.3}$ at room temperature and 500 °C.

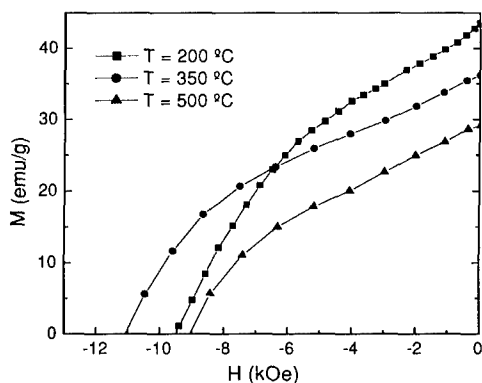


Figure 6. Demagnetization curves of $\text{SmCo}_{6.0}\text{Fe}_{0.4}\text{Cu}_{0.6}\text{Ti}_{0.3}$ at different temperatures.

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