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Development of high-energy product $Sm_2Fe_{17}N_3$ bonded magnets

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The $Sm_2Fe_{17}N_3$ has excellent intrinsic magnetic properties for permanent magnets. We have developed high performance bonded magnets; the $(BH)_{max}$ values of compression-molded and injection-molded bonded magnets are 20 and 13 MGOe, respectively. Here, we introduce the present situation and a view of our research on the $Sm_2Fe_{17}N_3$ magnets.

1. INTRODUCTION

High saturation magnetization of an iron nitride was reported by Kim and Takahashi in 1972 [1]. On the basis of the idea that it is necessary to make the iron nitride have a large magnetocrystalline anisotropy in order to attain a high coercivity, we started an investigation of structural and magnetic properties of Fe-N-*R* ternary system (where *R* denotes a rare earth element). In 1987, we found out a new compound $Sm_2Fe_{17}N_x$ which is a good candidate for permanent magnets [2].

The $Sm_2Fe_{17}N_x$ is a new type of magnet material which is prepared by introducing nitrogen atoms into the interstitial sites of the Sm_2Fe_{17} crystal under a nitrogen-containing atmosphere at elevated temperatures. The introduction of nitrogen atoms brings out the drastic changes in the intrinsic magnetic properties. The most excellent magnetic properties can be attained in the sample with x=3.0, the saturation magnetization being 15.7 kG, the anisotropy field 260 kOe and the Curie temperature 473 ℃ [3]. The value of the theoretical maximum energy product $(BH)_{max}$ amounts to 62 MGOe, being a match for the one of the $Nd_2Fe_{14}B$.

It has been found that the coercivity of the $Sm_2Fe_{17}N_3$ increases with decreasing size of the particle [4]. Utilizing these favorable characteristics, we have succeeded in developing high performance bonded-magnets. In this paper, we introduce the process of the fabrication of the $Sm_2Fe_{17}N_3$ bonded-magnets and the characteristics of the bonded magnets.

2. PRODUCTION OF $Sm_2Fe_{17}N_3$ BONDED-MAGNETS

2.1. Preparation of $Sm_2Fe_{17}N_x$ powder

The typical preparation step for the $Sm_2Fe_{17}N_x$ powder is shown in Figure 1. The host alloy Sm_2Fe_{17} is prepared by induction-melting Sm and Fe. The cast ingot is homogenized by annealing in an argon atmosphere at 1000 ~ 1250 ℃. The alloy is coarsely pulverized and then nitrogenated in a furnace with a mixed gas of NH_3 and H_2. The quantity of nitrogen uptake into the alloy powder varies with the conditions of nitrogenation. Conditions of nitrogenation and nitrogen content x of the $Sm_2Fe_{17}N_x$ powder are given in Table 1. It can be seen from the table that the nitrogen content x is changeable up to x = 6. The $Sm_2Fe_{17}N_x$ powder in which $x > 3.0$, however, do not consist of a single phase of the Th_2Zn_{17} structure [3]. According to the neutron diffraction study of $Nd_2Fe_{17}N_x$, the nitrogen atoms are located only in 9*e* sites which admit 3 atoms per formula unit [5]. This fact suggests that, in the $Sm_2Fe_{17}N_x$ powder in which $x > 3.0$, the $Sm_2Fe_{17}N_3$ phase coexists with an amorphous phase having higher nitrogen content. This coexistence causes the deterioration of the saturation magnetization. Annealing the $Sm_2Fe_{17}N_x$ powder in which $x > 3.0$ in an argon atmosphere, removes appreciable amounts of nitrogen, so that the nitrogen content x approaches 3.0. This annealing is very effectual in obtaining a $Sm_2Fe_{17}N_3$ powder which shows most excellent hard magnetic properties.

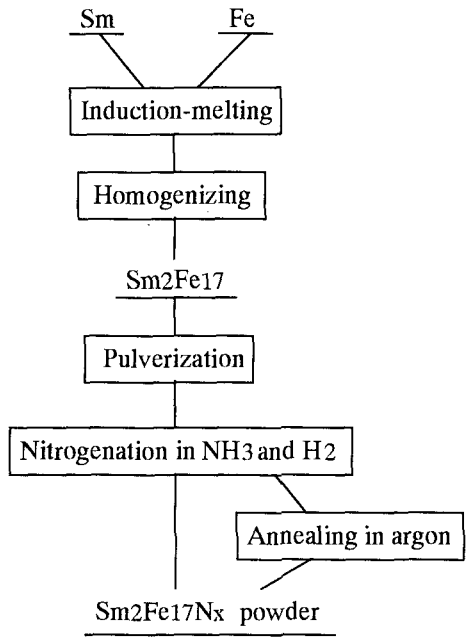

Figure 1. Typical preparation step for Sm2Fe17Nx.

2.2. Method of magnet fabrication

Although coarse powder shows low coercivity, fine powder with a 2 ~ 3 μm size has high coercivity [4]. Therefore, bonded-magnets can be fabricated by mixing the fine powder with a polymer or metal binder, and then aligning and compacting the compounds. Figure 2 shows the flow charts of the fabrication of compression-molded and injection-molded magnets and zinc-bonded magnets.

3. CHARACTERISTICS OF $Sm_2Fe_{17}N_3$ BONDED-MAGNETS

3.1. Magnetic properties of $Sm_2Fe_{17}N_3$ bonded magnets

Typical examples of the demagnetization curves and magnetic properties of the bonded-magnets are shown in Figure 3 and Table 2, respectively. It is notable that both compression- and injection-molded magnets have higher $(BH)_{max}$ values than commercial magnets, the $(BH)_{max}$ values of the commercial Sm-Co compression-molded magnets being 15 ~ 17 MGOe, Sm-Co injection-molded magnets 10 MGOe and Nd-Fe-B compression-molded magnets 10 MGOe. The injection-molded magnet has relatively low coercivity, since the deterioration of the coerciv-

Table 1
Condition of nitrogenation and nitrogen content x in Sm2Fe17Nx powder

Sample name	Condition of nitrogenation				nitrogen content x in Sm2Fe17Nx
	Temperature (℃)	Partial pressure (atm) NH3	H2	Time of nitrogenation (min)	
a	420	0.35	0.65	1	0.9
b	465	0.35	0.65	1	1.9
c	465	0.35	0.65	15	2.8
d	465	0.35	0.65	120*	3.0
e	465	0.35	0.65	120	3.3
f	465	0.35	0.65	960	4.1
g	495	0.35	0.65	120	4.8
h	465	0.45	0.55	150	5.9

* Sample d was heated in argon atmosphere for 1 hour at 465 ℃ after nitrogenation to reduce nitrogen content.

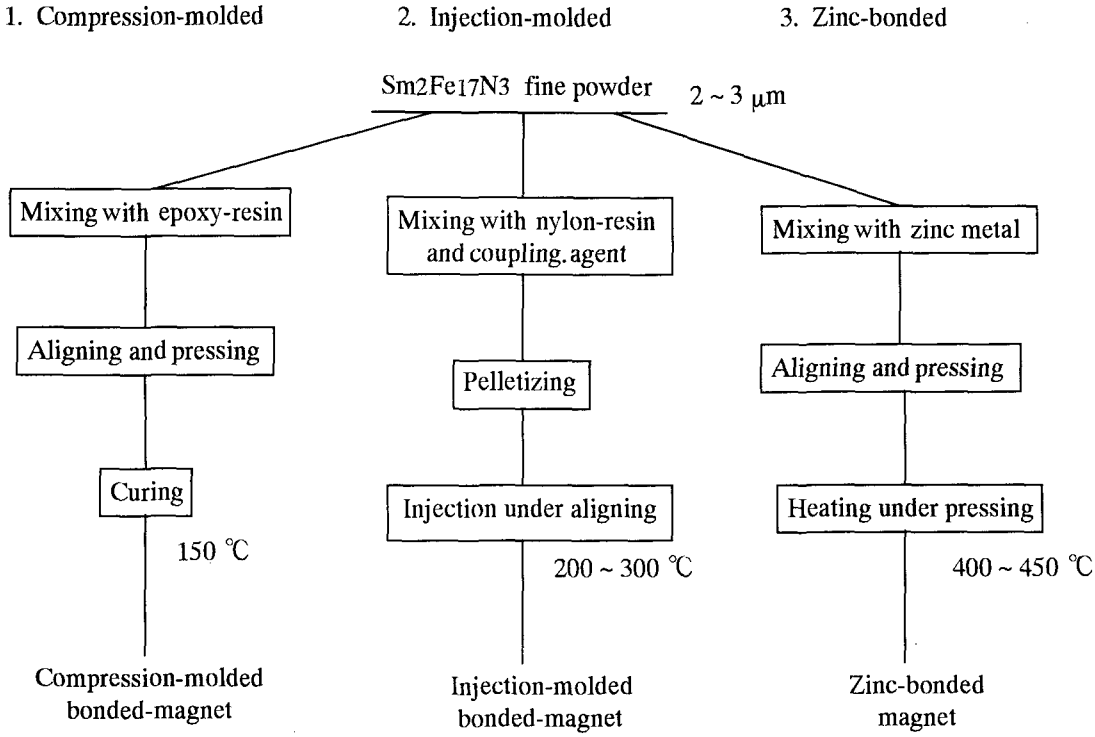

Figure 2. Flow charts of the fabrication of compression-molded and injection-molded magnets and zinc-bonded magnets.

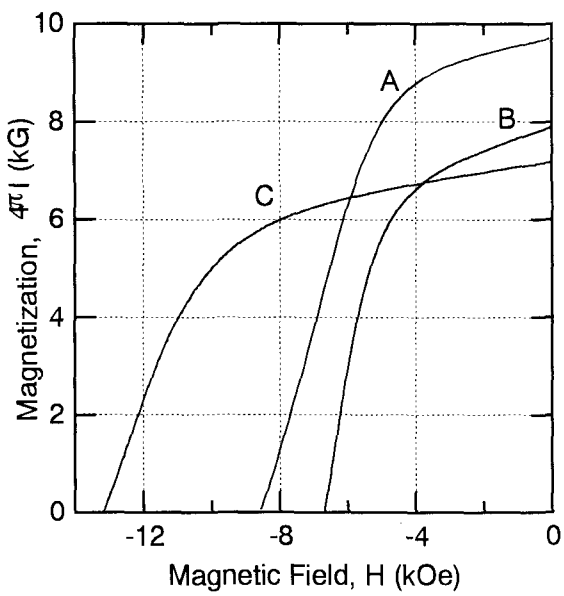

Figure 3. Demagnetization curves of various $Sm_2Fe_{17}N_3$ bonded- magnets. (A: epoxy-bonded compression, B: nylon-bonded injection, C: zinc-bonded)

ity occurs during the injection process due to high temperature. In contrast, the zinc-bonded magnet has high coercivity resulting from a heat treatment around the melting point of zinc. It is supposed that this enhancement of coercivity is owing to the emergence of Zn_7Fe_3 [6] or $Sm_2(Fe_{0.7}Zn_{0.3})_{17}N_3$ phase [7].

3.2. Thermal properties and oxidation resistance

Thermal properties of $Sm_2Fe_{17}N_3$ bonded-magnets are shown in Table 3. Although temperature coefficients of remanence (α) of $Sm_2Fe_{17}N_3$ bonded-magnets are fairly good, temperature coefficients of coercivity (β) of epoxy and nylon bonded-magnets are inferior to the other rare-earth magnets. The addition of cobalt raises the Curie temperature (554 ℃), but has no effect on the temperature coefficients α and β. Figure 4 shows the irreversible flux loss of injection-molded magnets (L/D = 0.7). It can be seen that the addition of cobalt is effective for the improvement of the irreversible flux loss.

Table 2
Magnetic properties of $Sm_2Fe_{17}N_3$ bonded-magnets

Magnet	Density (g/cm^3)	Remanence, Br (kG)	Coercivity, iHc (kOe)	(BH)max (MGOe)
Compression-molded (Epoxy-bonded)	5.8 ~ 6.0	9.5 ~ 10.0	7 ~ 9	18 ~ 20
Injection-molded (Nylon-bonded)	4.5 ~ 5.0	7.5 ~ 8.0	5 ~ 7	12 ~ 13
Zinc-bonded	7.2 ~ 7.5	7.0 ~ 9.5	7 ~ 15	10 ~ 17

Table 3
Thermal properties of $Sm_2Fe_{17}N_3$ bonded-magnets

Magnet	Curie temperature (℃)	Temperature coefficient (%/degree)	
		Remanence, α	Coercivity, β
Compression-molded (Epoxy-bonded)	473	− 0.06 ~− 0.085	− 0.45
Injection-molded (Nylon-bonded)	473	− 0.06 ~− 0.085	− 0.45
Zinc-bonded	473	− 0.06 ~− 0.085	− 0.35
$Sm_2(Fe_{0.9}Co_{0.1})_{17}N_3$ Injection-molded (Nylon-bonded)	554	− 0.06 ~− 0.10	− 0.45

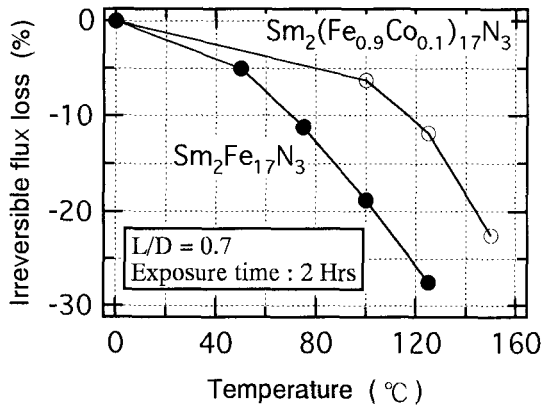

Figure 4. Irreversible flux loss of injection-molded magnets.

Since $Sm_2Fe_{17}N_3$ bonded-magnets comprise fine powder, we must be concerned with preventing from an oxidation of the powder. A polymer-bonding method is favorable for attaining an excellent oxidation-resistance. A nylon-bonded magnet has a fairly good oxidation resistance by the coating the fine powder with the nylon. Some spots of rust, however, can be observed after the humidity test (80 ℃, 90 %RH, 500 hrs). It has been found that the oxidation resistance can be improved with the addition of cobalt; no spots of rust can be observed after the same humidity test of the $Sm_2(Fe_{0.9}Co_{0.1})_{17}N_3$ nylon-bonded magnet. The addition of cobalt has a good effect on not only the irreversible flux loss but also the oxidation resistance.

5. SUMMARY

The Sm-Fe-N compounds are highly favorable to its utilization as the materials for permanent magnet applications, especially for bonded magnets. The $(BH)_{max}$ values of $Sm_2Fe_{17}N_3$ compression-molded and injection-molded magnets are 20 and 13 MGOe, respectively. Although some problems such as the relatively large irreversible flux loss remain for practical use, it seems that these problems will be cleared up by further studies for improvement.

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