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MAGNETIC AFTEREFFECT IN RARE EARTH-IRON-BORON MAGNETS

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ABSTRACT

The temperature dependences of the aftereffect coefficient Sv and the coercive force iHc have been measured from 4.2K to 300K on two specimens prepared from sintered magnets of $Pr_8Y_7Fe_{77}B_8$ (sintered at 1060 °C and 1100°C). The latter has higher maximum energy products. The Sv values of both have a maximum at 60K and 150K respectively. This is a new behavior which can not be explained by any theory proposed until now.

INTRODUCTION

If a certain magnitude of an external magnetic field is applied to a ferromagnetic specimen suddenly, in general, the consequent change of the intensity of magnetization is not correspondingly rapid. For all ferromagnetic substances there is a time lag between the magnetic field and the magnetization which is not caused by the eddy current contribution. This phenomenon is now generally called "magnetic viscosity" or "magnetic aftereffect". The earliest observations of viscous magnetization were made on Iron by Ewing [1] , and then many works have been accumulated on this phenomenon. Later, the phenomenon was classified into the two categories by Néel [2-4], namely:

1. Diffusion aftereffect
2. Thermal fluctuation aftereffect

The former is related with the diffusion of impurities, for instance, Carbon or Nitrogen atoms in a pure Fe crystal, and the latter is induced by the thermal fluctuation and is found more or less in all ferromagnetic materials. It is found [5] that the thermal fluctuation aftereffect is represented by the equation:

$$\Delta I'=\chi'_{irr}Sv(\ln t_2-\ln t_1)/(1-N\chi'_{rev}) \qquad (1)$$

where $\Delta I'$ is the difference of the apparent magnetization (namely, without correction for a demagneting field) between t_1 and t_2 which are the times of measurement after changing the field, χ'_{irr} an apparent irreversible differential susceptibility, χ'_{rev} the apparent reversible susceptibility and N the demagnetizing factor of the specimen. In general, the time associated with eddy current effects is very small, so it is negligible.

The thermal fluctuation aftereffect appears remarkably strong in magnetically hard materials, especially in rare earth magnets. Recently, much attention has been focused on the R-Fe-B material (where R is rare earth elements), since high energy permanent magnets having maximum energy products larger than 280 KJ/m^3 (35MGOe) were developed on the basis of the intermetallic compound $Nd_2Fe_{14}B$. From the point of view of the practical use, the existence of a large magnetic aftereffect is an undesirable characteristic for the permanent magnets. We have studied the thermal fluctuation aftereffect in R-Fe-B magnets, especially in $Pr_8Y_7Fe_{77}B_8$ magnets, and determined the thermal fluctuation aftereffect coefficient Sv in the neighborhood of the coercive force as a function of temperature, because in this region the thermal fluctuation aftereffect appears most pronounced.

EXPERIMENTAL PROCEDURE

Two specimens were sintered at 1100°C and 1060°C respectively. In this paper, the specimen sintered at 1100°C will be called as specimen Ⅰ and the other specimen Ⅱ. Both of them were formed into spheres of about 5mm in diameter. Aftereffect measurements were carried out at temperatures between 4.2K and 300K in external fields up to 5.2T, using a superconducting magnet. Magnetization values of specimens were determined by the induction method using a precision digital magnetometer which can resolve a relative magnetization change of 5×10^{-5} . The rate of the field change is 15600 A/m per second.

In a region of the hysteresis loop which is close to the coercive field, and shown in Fig.1, the magnetization I_A is measured at $H=H_0$. This measurement is made 30 seconds after H reached to H_0. This interval of 30 seconds is necessary for the field to be stabilized. Then the magnetic field is changed to a new point H_1, and the difference of the magnetization is measured as $\Delta I'=I_B-I_C$ from 30 seconds to 10 minutes after the magnetic field reaches H_1 . At last, the magnetic field is returned to H_0 again, the magnetization is measured as I_D 30 seconds after the magnetic field reaches H_0. From eq.(1), the aftereffect coefficient Sv can be obtained by substituting the following relations: $\chi'_{irr}=(I_A-I_B)-(I_D-I_C)$, $\Delta I'=I_B-I_C$, $\chi'_{rev}=I_D-I_C$.

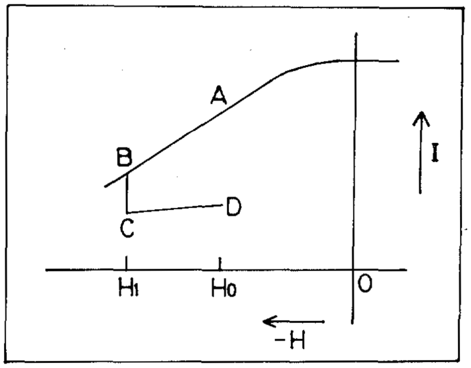

Fig. 1 The experimental procedure for measuring the aftereffect coefficient Sv.

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RESULTS AND DISCUSSION

An example of the aftereffect observed at 60K on specimen II is shown in Fig.2. From this figure we can find out that the results of our experiment accord with the equation (1) very well. For the investigated specimens, the total variation of magnetization during the time interval of observation is small; its order of magnitude is about 1 percent of the saturation magnetization. Figure 3 shows the measured Sv values as a function of temperature for both specimens. The coercive force changes with temperature as shown in Fig.4 and the apparent irreversible differential susceptibility changes with temperature as shown in Fig.5. It is a very interesting fact that the Sv value of the specimen II has a maximum at about 60K, which corresponds exactly to the case of $Nd_{15}Fe_{77}B_8$ magnet[6], and which is quite contrary to the case of $SmCo_5$ magnets where Sv increases with increasing temperature(cf Fig.6)[5], almost following the Néel law, $Sv \sim T^{1/2}$ [11] The Sv value of the specimen I has a maximum at about 150K, the peak is very broad and the magnitude larger than that of the specimen II The coercive force iHc of both specimens decreases with increasing temperature, but the magnitude of the coercive force of the

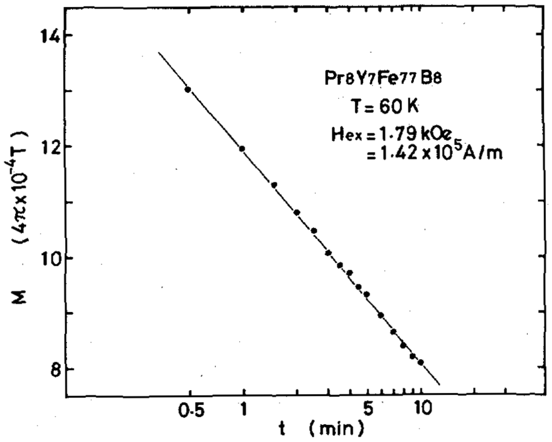

Fig. 2 An example of the aftereffect observed at 60K on specimen II.

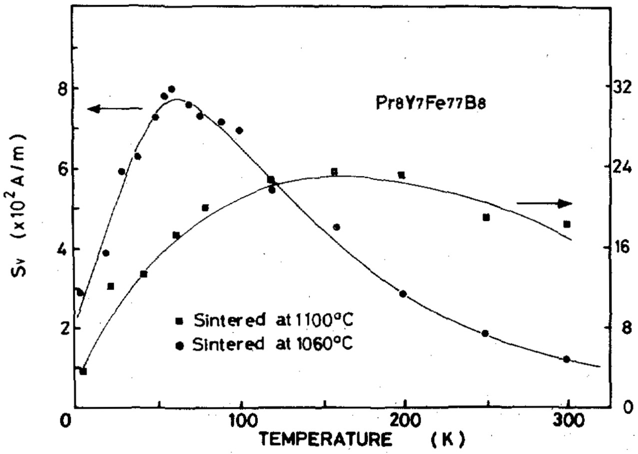

Fig. 3 Temperature dependence of the aftereffect coefficient Sv.

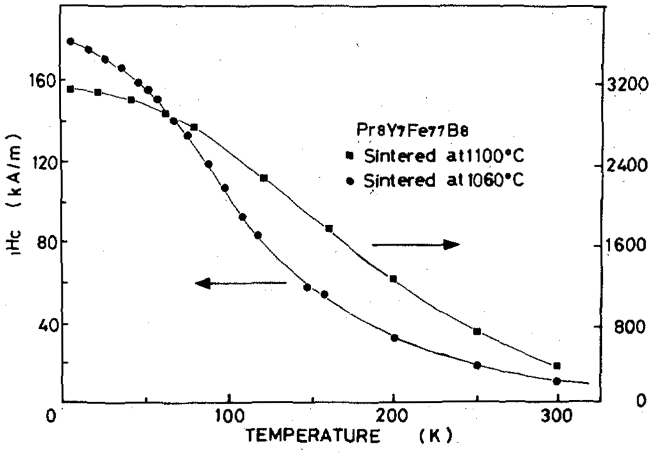

Fig. 4 Temperature dependence of the coercive force iHc.

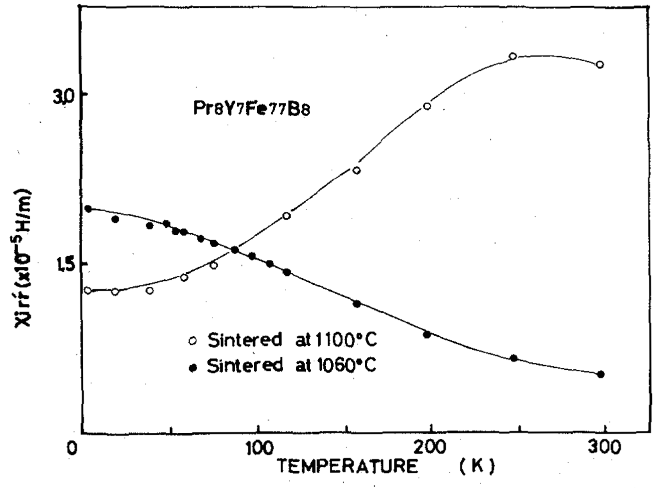

Fig. 5 Temperature dependence of the apparent irreversible differential susceptibility.

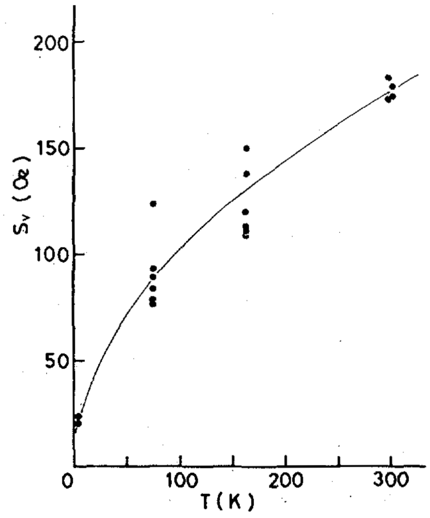

Fig. 6 Temperature dependence of the aftereffect constant Sv for a SmCo5 magnet[5].

specimen I is much larger than that of specimen II. The behavior of the apparent irreversible differential susceptibility χ'_{irr} between the two specimens is quite different: The χ'_{irr} of the specimen I increases and that of the other decreases with increasing temperature.

About the relation between the viscosity coefficient Sv and the temperature, there have been many investigations, but their results are not consistent. For example, on Alnico $Sv \sim T^{3/4}$ was found by Barbier[7], $\chi_{irr}Sv \sim T$ by Street and Wooley[8], $Sv \sim T^{3/2}$ by Matsuura[9]; on Ni_3Mn $\chi_{irr}Sv \sim T$ by Taoka[10]; and on $SmCo_5$, $Sv \sim T^{1/2}$ by Yamada[5]. On the theory side, there are also different conclusions. Some of them which are important are the following :

$$Sv^2=4\pi kT/6V(Q'+\ln\tau) \qquad (2)$$

which was derived by Néel [11], and

$$S=\chi_{irr}Sv=NMs\lambda kT \qquad (3)$$

by Brown [12], where Ms is the spontaneous magnetization, V the activation volume swept out the energy barrier, kT the Boltzmann energy, τ the "waiting time" before a barrier is overcome at an absolute temperature T, N the number of the particles which possess the "volume" V, λ the value that determine the distribution of the particles which possess an anisotropy energy W, and Q' a constant.

From the discussion made above, we can say that the results of our experiment reveal a new behavior in the temperature dependence of the viscosity coefficient, that is not explained by any existing theory. From Figs.3-5, we can also see that the differences of the properties between the two specimens are quite large, although the two specimens have the same chemical composition. So we can say that Sv contains detailed information on the elementary magnetization process and is sensitive to the micromechanism of the specimen. Because the magnetic aftereffect is due to a great number of complexly correlated magnetic processes, the effects can hardly be described in simple terms.

Finally it is interesting to compare the temperature dependence of the aftereffect coefficient Sv of the investigated R-Fe-B magnets to that of $SmCo_5$, because both are classified as the nucleation controlled type, apparently distinguished from the pinning type such as Sm_2Co_{17}-based magnets. The temperature dependence of the aftereffect coefficient Sv of the $SmCo_5$ sintered magnets is shown in Fig.6 [5]. At low temperature, both magnets show a similar trend of increasing Sv. However, at higher temperature a distinct difference emerges between R-Fe-B and $SmCo_5$. Namely, in the latter, Sv still increases with temperature, whereas in the former, the Sv decreases with temperature. In order to explain this behavior, we have to consider the following facts [13]: In $SmCo_5$ magnet, the Curie temperature of the grain boundary region is higher than in the matrix phase. Therefore, a relatively high value of magnetic anisotropy is maintained at temperatures close to Tc of the matrix. Contrarily, in the R-Fe-B magnets the grain boundary regions (Nd-rich phase and B-rich phase) are always magnetically softer than the matrix phase because of its negligible anisotropy and low Tc. Now, let us suppose the following: the difference in the magnetic properties of the grain boundary phases between the investigated R-Fe-B and $SmCo_5$ permanent magnets makes the temperature dependence of the activation volume V of eq.(2) different. In the case of R-Fe-B magnet, V changes with increasing temperature, while in the $SmCo_5$, V remains unchanged as supposed by Néel. So it appears that the existence of the boundary phase in the grain boundary region of the sintered R-Fe-B magnets is an essential factor in the magnetic aftereffect mechanism in this type of magnet.

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