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STUDY OF REVERSIBLE AND IRREVERSIBLE MAGNETIZATION COMPONENTS IN HYBRID Nd-Fe-B /FERRITE/ ALNICO RESIN BONDED MAGNETS

BADANIE ODWRACALNEJ I NIEODWRACALNEJ SKŁADOWEJ NAMAGNESOWANIA W HYBRYDOWYCH MAGNESACH Nd-Fe-B/FERYT/ALNICO WIĄZANYCH TWORZYWEM SZTUCZNYM

Magnetic properties of hybrid resin bonded magnets produced from a mixture of MQP-B with strontium ferrite or alnico ones have been investigated. The room temperature magnetic measurements as the initial magnetization curve, the major and minor hysteresis loops and two sets of recoil curves have been performed. The data from all recoil loops have been used to derive the field dependence of the reversible and irreversible magnetization components of the total magnetization. From both the initial and demagnetization curves the field dependence of the differential susceptibility were received. It was found from that the main mechanism of magnetization reversal process in MQP-B and MQP-B with strontium ferrite magnets is the pinning of domain walls. For the MQP-B magnet with alnico apart from the pinning of domain walls at the boundaries of MQP-B grains, the rotation of the magnetization vector in alnico grains contributes mainly to the reversible magnetization. The interactions have been studied by means of the Henkel plots. In bonded magnets made from MQP-B and MQP-B with alnico the dipole interactions are dominant while in MQP-B with strontium ferrite magnet the s-shaped behaviour of the Henkel plots have been observed as in exchange-coupled magnets.

Keywords: HYBRID MAGNETS, INTERACTION DOMAINS, PINNING OF DOMAIN WALLS, MAGNETIZATION REVERSAL PACS: 71.20 Be; 74.25. Ha; 75.50. Tt; 75.60. -d; 75.60 Jk

W pracy badano własności magnetyczne hybrydowych magnesów wiązanych żywicą, wyprodukowanych z mieszaniny proszków MQP-B i ferrytu strontu lub alnico. Pomiary magnetyczne takie jak: pierwotne krzywe magnesowania, główne i częściowe pętle histerezy oraz krzywe powrotne przeprowadzono za pomocą magnetometru wibracyjnego LakeShore w polu magnetycznym do 2T i magnetometru Bittera w polu do 14 T. Dane z pomiaru wszystkich krzywych powrotnych posłużyły do wyznaczania zależności składowej odwracalnej i nieodwracalnej namagnesowania od wartości przyłożonego pola magnetycznego. Z krzywych pierwotnych i krzywych odmagnesowania wyznaczono zależności różniczkowej podatności jako funkcji przyłożonego pola magnetycznego. Z tych zależności wynika, że głównym mechanizmem procesu przemagnesowania w magnesach otrzymanych z proszku MQP-B i mieszaniny proszku MQP-B z ferrytem strontu jest kotwiczenie ścian domenowych na granicach ziaren MQP-B. Natomiast w magnesie z dodatkiem alnico oprócz kotwiczenia ścian domenowych występują obroty wektora namagnesowania w ziarnach alnico. Z zależności Henkla wynika, że w magnesach z czystego MQP-B i MQP-B z dodatkiem alnico dominują oddziaływania dipolowe. Natomiast dla magnesu z dodatkiem ferrytu strontu krzywa Henkla ma przebieg charakterystyczny dla magnesów, w których dominuje sprzężenie wymienne.

1. Introduction

Bonded permanent magnets are now produced by many manufacturers. They find still growing application in the industry and technology owing to many advantages in comparison with sintered magnets. Even though the worse magnetic properties the bonded magnets have better mechanical properties, lower costs of production and possibility to obtain the different shapes with very high accuracy.

The bonded magnets are produced from a mixture of hard magnetic powder with different kinds of polymers or resins. The magnetic properties of bonded magnets depend on the kind and volume fraction of the hard magnetic components, the kind and volume fraction of the binder and used technology [1-3]. In order to obtain the magnet with the properties desired in a given application the hybrid magnets are made from a mixture of two different hard magnetic powders. Magnetic properties of such magnets result not only from the kind of component

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but from the interaction between them as well. Therefore the hybrid bonded magnets are interesting for scientists giving them an opportunity to study the influence of different addition on this interactions and mechanisms of magnetization reversal processes and thus the magnetic properties.

The mechanisms occurring in the magnetization and demagnetization processes in nanocrystalline magnets consist of the reversible and irreversible ones. The rotations of the magnetization vector in single domain particle of the melt quenched materials and bowing of the domain walls pinned at the grain boundaries are the processes which contribute to reversible magnetization. However, the pinning of domain walls at the grain boundaries which were found to be the thin layer of amorphous phase (some nm thickness) is the main mechanism of the irreversible magnetization reversal.

The aim of this paper was the study of the alnico and ferrite addition on the magnetization reversal processes in bonded magnets made from nanocrystalline Nd-Fe-B material.

2. Experimental details

The magnets investigated have been produced using compression-moulding technique at a pressure of 900 MPa and cured at temperature of 180°C for 2 hours. The starting materials for preparing the samples were composed of 100 wt % of Magnequench MQP-B commercial powder [4] obtained from the melt quenched Nd-Fe-Co-B nanocrystalline ribbons with the average grain size of 163 nm (sample a), a mixture of 70 wt % of MQP-B and 30 wt % of alnico powder (sample b) or 30 wt % of strontium ferrite powder (sample c). The Nd-Fe-Co-B ribbons were ground into particles with average size ~200 μm. For alnico and ferrite powders these sizes are 250 μm and 50 μm, respectively. 2.5 wt % of epoxy resin has been used as a binder [1].

The magnetic measurements have been performed at room temperature on cylindrical samples with the Lake Shore and Bitter vibrating sample magnetometers with the maximum applied magnetic field of 2 T and 14 T, respectively. The magnetic measurements data have been corrected for the demagnetizing field according to the sample geometry. The magnetic studies included the major hysteresis loops with initial magnetization curves, the series of minor hysteresis loops, the sets of recoil curves measured from different points on the initial magnetization and demagnetization curves to zero internal field. In order to record the recoil loops, a certain magnetizing (demagnetizing) field μ_0H_i ($-\mu_0H_i$) was applied to the sample in a demagnetized state (the remanent state after magnetizing by a field of 14 T). Next the field was decreased to zero and then increased to μ_0H_i+0,05 T ($-\mu_0H_i$-0,05 T). Such measurements have been done for 40 different values of μ_0H_i. From the recoil curves the reversible μ_0M_{rev} and irreversible μ_0M_{irr} components of the total magnetization μ_0M as a function of internal magnetic field have been derived. The irreversible magnetization component μ_0M_{irr} at a magnetizing (demagnetizing) field μ_0H_i ($-\mu_0H_i$) is defined as the magnetizing remanence $\mu_0M^r_{irr}$ (demagnetizing remanence $\mu_0M^d_{irr}$) when the magnetizing (demagnetizing) field μ_0H_i ($-\mu_0H_i$) is removed. The reversible component is equal to $\mu_0M_{rev} = \mu_0M - \mu_0M_{irr}$ [5-7].

By differentiating the $\mu_0M_{rev}(\mu_0H_i)$ and $\mu_0M_{irr}(\mu_0H_i)$ dependencies the reversible χ_{rev} and irreversible components χ_{irr} of total susceptibility as a function of internal field have been obtained.

The interaction between grains has been determined by studying the so-called Wohlfarth's remanence relationship [8]

$$M^d_{irr}(\mu_0H_i)/M_R = 1 - 2 \cdot M^r_{irr}(\mu_0H_i)/M_R \qquad (1)$$

According to Wohlfarth the dependence (1) is valid for noniteracting single domain particles therefore, each deviation from equation (1) can be interpreted as being due to the magnetic interaction between grains or magnet particles.

3. Results and discussion

Fig 1. shows the major hysteresis loops for three samples measured in a magnetizing field of 14 T. From these loops the magnetic parameters like the remanence μ_0M_R, the coercivity μ_0H_C and the maximum energy product $(BH)_{max}$ were determined and listed in Table 1.

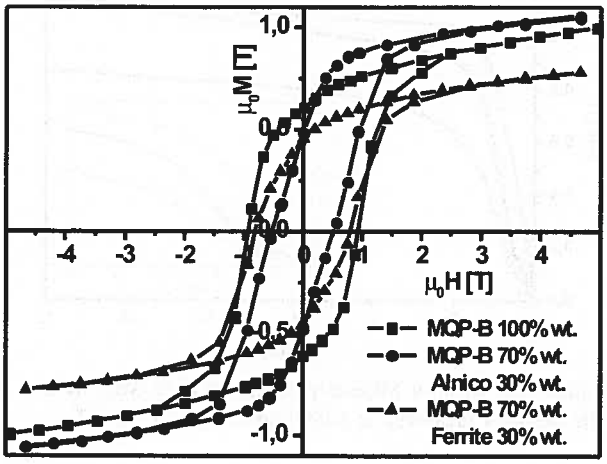

Fig. 1. Major hysteresis loops for the samples 100 wt.% of MQP-B powder, 70 wt.% MQP-B and 30 wt % of alnico powders, 70 wt. % of MQP-B and 30 wt. % of strontium ferrite powders

It is seen in Table 1 that the magnet with addition of alnico has the saturation magnetization almost the same as the magnet made from pure MQP-B powder and lower values of the coercivity, remanence and maximum energy product. The addition of strontium ferrite reduces all parameters but the value of the coercivity 0,80 T and remanence 0,47 T are higher than that of 0,6 T and 0,42 T, respectively, expected from a dilution law for the simple mixture of the MQP-B and strontium ferrite powders. On the other hand the changes of the parameters caused by the addition of alnico are as follows: the coercivity is higher and remanence lower (0,52 T and 0,48 T) than that resulting from the dilution law (0,42 T and 0,67 T, respectively). This implies that the changes result from both the mixing of the components with different magnetic properties and the interaction between the components particles.

TABLE 1

Magnetic characteristics of hybrid bonded Nd-Fe-B magnets with alnico or strontium ferrite powder content

Sample	$\mu_0 M_S$ [T]	$\mu_0 M_R$ [T]	$\mu_{0J} H_C$ [T]	$(BH)_{max}$ [kJ/m^3]
MQP-B 100% wg	1,12	0,61	0.93	57,5
MQP-B 70% wg Alnico 30% wg	1,14	0,48	0,52	23,7
MQP-B 70% wg strontium ferrite 30% wg	0,9	0,47	0,80	29,5

The initial magnetization curves measured in an external magnetic field up to 14T and 2T are presented in Fig 2a and in Fig 2b, respectively. The initial magnetization for sample "a" made from pure MQP-B powder increases slowly with increasing internal magnetic field up to about 0,7 T and then increases faster for higher field, approaching the saturation magnetization in a field of 12 T. Similar behaviour is seen for sample made from a mixture of MQP-B and ferrite powder (sample "c"). It is assumed that the pinning of domain walls is the main mechanism responsible for such behaviour of the initial magnetization curve. The initial curve for sample with alnico powder shows a steep rise in the magnetization as a result of a combination of two types of magnetization processes. For this sample, apart from the pinning of domain walls in MQP-B grains, the rotation of the magnetization vector out of the easy axis in alnico grains exists and plays the dominant role in magnetization process. The non-zero slope of the initial magnetization curves at low fields indicates that another mechanism is taking part yet in magnetization process. This is the rotation of the magnetization vector in MQP-B single domain grains.

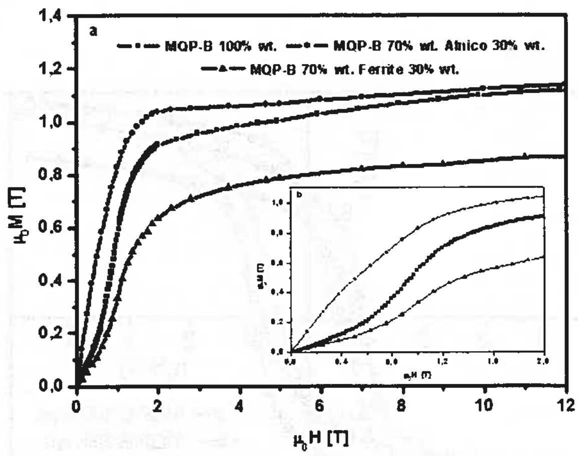

Fig. 2. Initial magnetization curves for samples 100 wt.% of MQP-B powder, 70 wt.% MQP-B and 30 wt % of alnico powders, 70 wt. % of MQP-B and 30 wt. % of strontium ferrite powders measured at a field up to (a)14 T, (b) 2T

Fig. 3 presents the dependence of the coercivity (a), and remanence (b) derived from the minor hysteresis loops on the maximum applied magnetizing field. The slow increase in both parameters at low internal fields is followed by their faster increase in higher fields except for the sample with strontium ferrite for which the parameters initially develop more rapidly, indicating the existence of pinning sites in ferrite grains with pinning fields lower than that one in MQP-B grains. The coercivity and the remanence of all the samples reach the saturation value when the internal field is over 3T. Such behaviour indicates that with increasing field more and more domain walls are removed from the pinning sites.

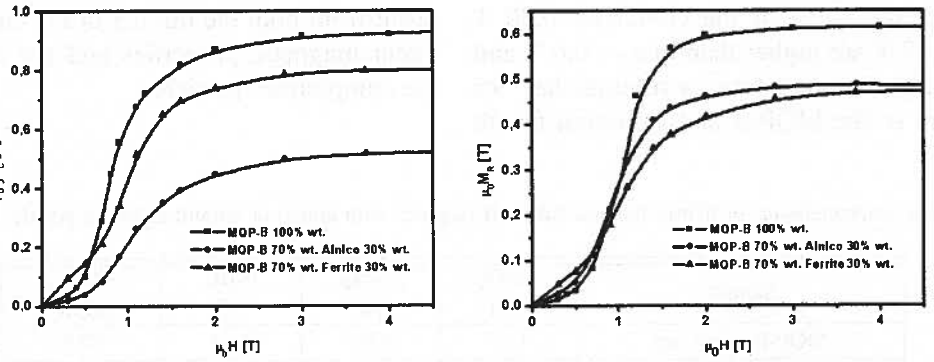

Fig. 3. Dependence of the a) coercivity b) remanence for the samples 100 wt.% of MQP-B powder, 70 wt.% MQP-B and 30 wt % of alnico powders, 70 wt. % of MQP-B and 30 wt. % of strontium ferrite powders on the maximum magnetizing field

More details about magnetization processes can be obtained by studying the recoil loops presented in Fig.4a-c. For the sample "a" and "c" they show a similar degree of reversibility in magnetization and demagnetization direction whicht means that the same processes contribute to reversible magnetization components. These processes are the rotation of the magnetization vector in single domain MQP-B grains and domain walls bowing. The recoil curves for the sample "b" with alnico show a higher degree of reversibility which originates from the reversible rotation of the magnetization vector in alnico grains. The contribution of the reversible and irreversible processes into magnetization and demagnetization is seen in Figs. 5 a-c and 6 a-c which present the field dependence of the reversible and irreversible magnetization and differential susceptibility components during the initial magnetization and demagnetization for the three magnets investigated. The reversible components in both direction change in a similar way for the sample "a" and "c". Initially they increase faster and remain greater than those corresponding with the irreversible components up to value of internal field close to the sample coercivity. In the sample "b" the reversible component predominates the irreversible one in the whole range of internal fields which means that the rotation of the magnetization vector in alnico grains gives the main contribution to the magnetization and demagnetization processes.

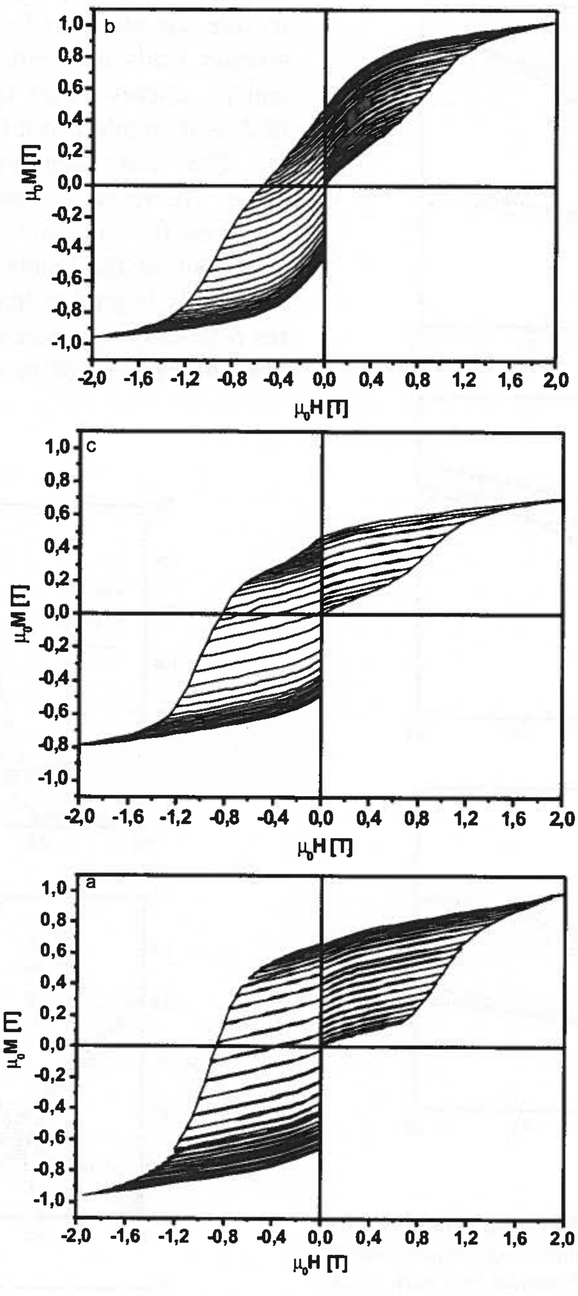

Fig. 4. Recoil curves for the samples a) 100 wt.% of MQP-B powder, b) 70 wt.% MQP-B and 30 wt % of alnico powders, c) 70 wt. % of MQP-B and 30 wt. % of strontium ferrite powders measured from different points on the initial magnetization curve and the demagnetization curve

As shown in Figs. 5 the shapes of the irreversible magnetization components dependence versus an internal field are different in forward and backward direction but in contrast to the sample "a" and "b" for the sample "c" the irreversible magnetization component reaches larger values in forward direction in comparison with the backward one. This may be attributed to the different character of intergrain interaction.

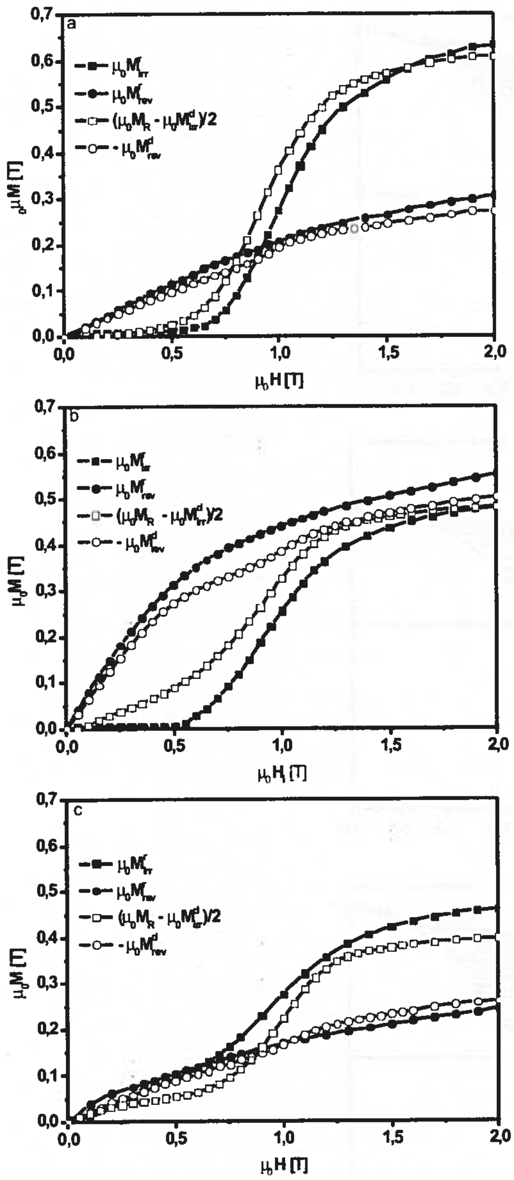

Fig. 5. Reversible $\mu_0 M\mu_{rev}$ and irreversible $\mu_0 M^r_{irr}$ components of the total magnetization $\mu_0 M$ during the initial magnetization and the demagnetization processes as a function of internal field $\mu_0 H_i$ (a) 100 wt.% of MQP-B powder, (b) 70 wt.% MQP-B and 30 wt % of alnico powders, (c) 70 wt. % of MQP-B and 30 wt. % of strontium ferrite powders

The changes in magnetization are better visible in the field dependence of the differential susceptibility (Figs.6 a-c). The irreversible susceptibilities in both directions have their maxima near the coercivity of the sample made from MQP-B powder. This implies that the main magnetization reversal mechanism is the pinning of domain walls in MQP-B grains which are very fine and smaller than the single domain particle size. The observed in such materials domain structure extends over many grains and is called interaction domains [9,10]. The $\chi_{irr}(\mu_0 H_{-i})$ dependencies reflect the distribution of coercivities and are rather broad. The second maximum in magnetizing χ^r_{irr} and demagnetizing χ^d_{irr} at low field for the sample with ferrite arises from the pinning of domain walls in ferrite grains. The amplitudes of χ^d_{irr} and χ^r_{irr} curves for all samples do not differ by a factor of 2 as it should result from differentiating the equation (1). This is an additional proof of the interaction existence. The reversal components of the susceptibility are small for the sample "a" and "c" in the whole range of fields but for the sample with alnico the value of χ_{rev} at low fields is greater that that of χ_{irr} which proves that the rotations of magnetization vector in alnico dominates over the irreversible processes.

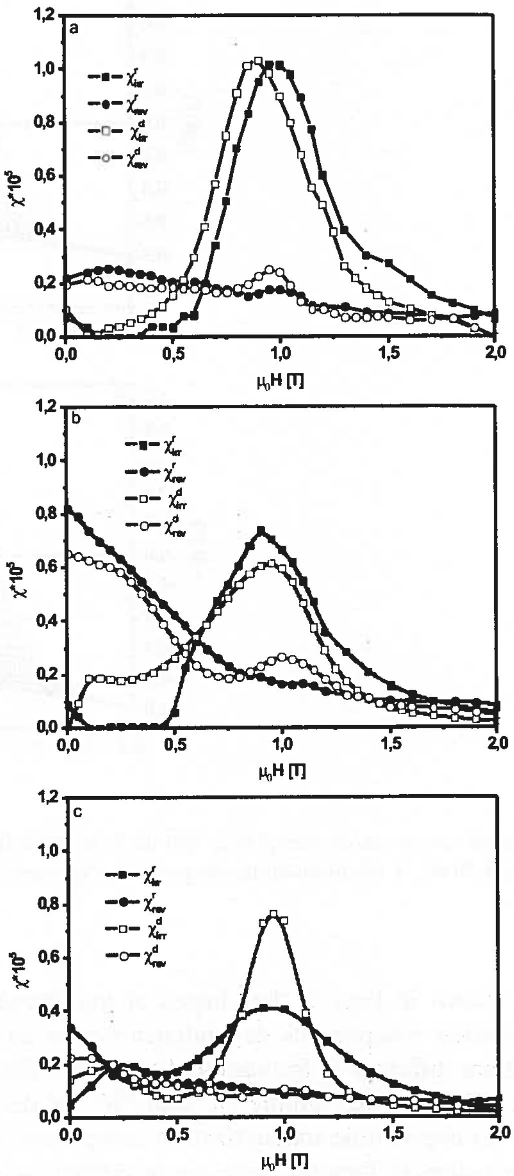

Fig. 6. Reversible χ_{rev} and irreversible χ_{irr} components of a total total susceptibility during the initial magnetization and the demagnetization processes as a function of internal field $\mu_0 H_i$ derived for the

sample made from pure MQP-B powder (a), 70 wt. % of MQP-B and 30 wt. % of alnico powders (b), 70 wt. % of MQP-B and 30 wt. % of strontium ferrite powders(c)

The interaction between grains can be studied by the so-called remanence relationship given by the equation (1). For the three samples investigated the relation is not obeyed and all the experimental points lie below the straight line for the sample "a" and "b" as it is seen in Fig. 7. This indicates that there exist interactions between the MQP-B particles, which are due to the strong magnetostatic coupling. The addition of alnico causes this interaction to be weakened because the sample particles are better separated. The magnet with strontium ferrite addition shows an S-like shape behaviour which is often observed in exchange spring magnets and suggests that in this sample the exchange coupling is dominant.

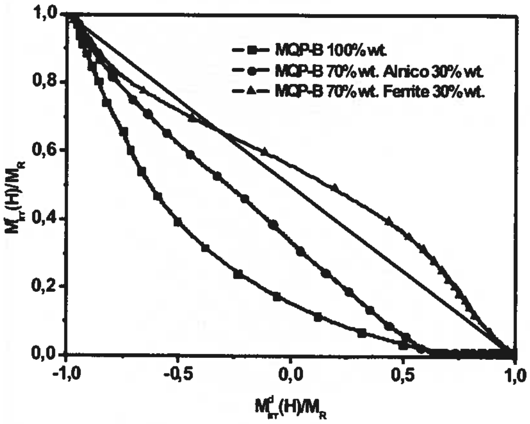

Fig. 7. Remanence relationship determined from recoil curves for the samples of 100 wt.% of MQP-B powder, 70 wt.% MQP-B and 30 wt % of alnico powders, 70 wt. % of MQP-B and 30 wt. % of strontium ferrite powders

4. Conclusion

The addition of alnico or strontium ferrite powders causes a decrease of the coercivity, remanence and maximum energy product values in comparison with the sample made from pure MQP-B powder. From the shape of initial magnetization curves at low fields and differential susceptibility it can be concluded that the main mechanism of magnetization reversal is the pinning of domain walls at the MQP-B grain boundaries in all the samples studied. In the sample with alnico addition the initial magnetization behaviour is a combination of the rotation of the magnetization vector in alnico grains and pinning of domain walls in MQP-B grains. From the dependence of the reversible and irreversible magnetization components as a function of the internal field magnetic field it results that the reversible changes of magnetization such as the rotation of magnetization vector in alnico and bowing of pinned domain walls in MQP-B grains have a higher influence on magnetization processes. Ferrite-hybrid magnet and pure MQP-B bonded magnet are dominated by pinning of domain walls that is by irreversible changes of magnetization.

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