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COMPOUNDING OF POLYMER BONDED Nd-Fe-B MAGNETS

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ABSTRACT

Polymer bonded Nd-Fe-B magnets belong to the group of functional composite materials, which consist mainly of two components: a polymer binder (PPS, PA) and magnetic particles of Nd-Fe-B alloy. The appropriate amounts of polymer binder, magnetic powder and additives are compounded by the twin screw extruder into a compound, that can be then injection moulded into a multi-cavity die in the final products of complex shapes in large series. To achieve a successful injection moulding and magnetic properties of the final products, it is necessary to choose the appropriate components for preparation of compound for polymer bonded magnets. To accomplish this the following parameters are crucial: the magnetic properties of powders, their fraction in the composite, shape, average size and size distribution of the particles, the particle-particle interaction, and the addition of additives (surface modifiers) and lubricants that improve the rheological, mechanical and corrosion properties of polymer bonded magnets.

Within the presentation we will discuss about the relations between: (i) optimal components fraction, (ii) magnetic and mechanical properties as well as (iii) acceptable viscosity of polymer melt during the injection moulding.

1. INTRODUCTION

Nowadays, in the information age, the polymer bonded magnets play a vital role as components in a wide range of devices (motors, sensors, actuators, acoustic transducers,). The demand in polymer bonded magnets is now the largest in the automotive, aviation industry and consumer electronics with an annual growth of sales in the global market of over 30%. Originally, in the 70's the polymer bonded magnets were made from ferrite powder and elastomers as a binder in a flexible form. To date, the powder may be also made from Nd-Fe-B or Sm-Co alloys and the binder form thermosets (epoxy) or thermoplastics (PA, PPS).

Polymer bonded magnets consist of at least two components a polymer binder (PPS, PA12) and magnetic particles of NdFeB alloy. From an appropriate mixture of polymer binder, magnetic powder and additives the granulated mixture is compounded by the twin screw extruder. The granulated mixture can be then injection moulded into a multi-cavity die in the final products of complex shapes in large series. Magnetic powders on the basis of NdFeB alloys with corresponding chemical composition are generally made by rapid solidification methods such as melt-spinning. Conventionally, alloys with nominal compositions near stoichiometric $Nd_2Fe_{14}B$ or $Nd_2Fe1_4B/\alpha Fe$ with small additions of elements like cobalt to maximized saturation magnetization are prepared by melt spinning [1]. In this method the liquid melt is ejected on the rapidly rotating drum, whereby, due to the large cooling rates (10^6K/s), which are the result of a large surface area of very thin strips of a thickness of some 10 μm, a completely amorphous or amorphous-nanocrystal microstructure is formed[1-3]. The controlled heat treatment to achieve nanostructure mono-domain microstructure with a grain size of approx. 40 to 50 nm is followed. After the heat treatment, the strips are crushed into the powder with a suitable size and, if necessary, sieved to achieve the appropriate particle size distribution. Due to the fine crystalline size the obtained ribbons are magnetically isotropic. For bonded magnets, powder magnetic properties, loading factor and moulding characteristics are important to obtain high maximum energy product $(BH)_{max}$ magnets. In general, powders with higher $(BH)_{max}$ are essential to achieve magnets oh high $(BH)_{max.}$ Additionally, the magnetic proprieties of bonded magnets also depend of powder morphology, type of binder, volume of pores and consolidation technique. In all cases, magnets would need to be moulded with commercially available moulding machines, which limit the filling ration of magnetic powder. Depending on type of polymer binder polyamides (PA6, PA12) or polyphenylene sulphide (PPS) the filling ratio for injection moulding can vary from 50 to 75 vol %. Usually, magnets with residual induction (Br), values of 480-600mT and $(BH)_{max}$ values of 40-55kJ/m3 can be obtained by injection moulding [3-4]. For high temperature applications the magnets powder and binder has to be considered. While PA binders are limited to a maximum of 120-150°C, on the other hand, the PPS allows higher operating temperature in the range of 180-200°C. One of important goal for magnet manufacturers is to develop net shape forming process to avoid and minimize machining costs. Fully dense sintered magnets are extremely brittle and so machining of such magnets is a cost effective and very demanding operation. On other hand, bonded magnets possess a high degree of net-shape formability and can be processed at relatively low temperatures (170-300°C) depending on the binder and moulding technique being used. Another advantage of injection moulding is the ability to mould onto other objects such as a shaft. Moreover, injection bonded magnets possess also excellent corrosion resistance, because during compounding each NdFeB particle is being covered in a protective polymer layer that preventing the powder to come into contact with atmosphere during operation.

This research work presents the results of development of polymer bonded Nd-FeB magnets by compounding on a twin screw extruder for application in automotive industry for fabrication of rotors for gasoline pumps. The investigation was made with collaboration with company Magneti Ljubjana d.d., Slovenia which is today a small but significant producer of permanent metallic magnets in Europe.

2. EXPERIMENTAL PROCEDURE

For compounding of granulate for polymer bonded magnets, the commercial melt spun powder (MQP 14-12, Magnequench) was selected as the magnetic powder. The MQP powder is a multimodal mixture of Nd-Fe-B particles having plate-like morphology with average particle size of 250 μm. It is specifically designed for high magnetic flux and high temperature application, such as automotive motors and sensors. This material is produced by employing a proprietary rapid solidification process followed by a milling process and heat treatment. The magnetic characteristics of the powder are: B_r=850 mT, $(BH)_{max}$ = 120 kJ/m^3 and H_{ci}= 1050 kA/m (H_{ci} -intrinsic coercivity) [5]. As a polymer binder the polyphenylene sulphide (PPS) from company Ticona was used in this study. The glass transition and melting temperature of the PPS was found to be 85°C and 285°C, respectively. To improve the processabillity, thermal oxidation resistance and mechanical properties of polymer bonded magnets, the MQP powder was treated with 3-Amino-propyl-tri-ethoxy Silane coupling agent by simple immersion of powders in aqueous solution of coupling agent. The excess solution was decanted and with Silane treated powder was dried in vacuum furnace at 80°C for 2 hours. Additionally, before mixing the PPS with MQP powder, the PPS was also dried to achieve constant weight. The PPS was dried four hours in a convection oven at 120°C. The desired amounts of MQP powder was dry mixed with PPS and other additives and lubricants in a Turbula Shaker-Mixer to achieved the homogenous mixture of magnetic and polymer powders. The Turbula Shaker-Mixer is specially designed for exact powder blending of extremely heavy powders with very light ones (ρ_{MQP}=7, 62 g/cm^3 and ρ_{PPS}=1, 36 g/cm^3). Finally, this mixture was compounded in a twin screw extruder (Leistritz ZSE 18HPe) and pelletized into cylindrical pieces of 2 mm diameter and 3 mm in length. Before injection moulding, the pelletized compound was dried in vacuum furnace at 120°C for two hours to assure moisture removal. The compound was than injection moulded (KraussMaffei CX 35-55) at 305°C to obtain test specimens in form of small disks (20 mm in diameter x 5 mm thick) for measurement of magnetic properties and metallographic examinations.

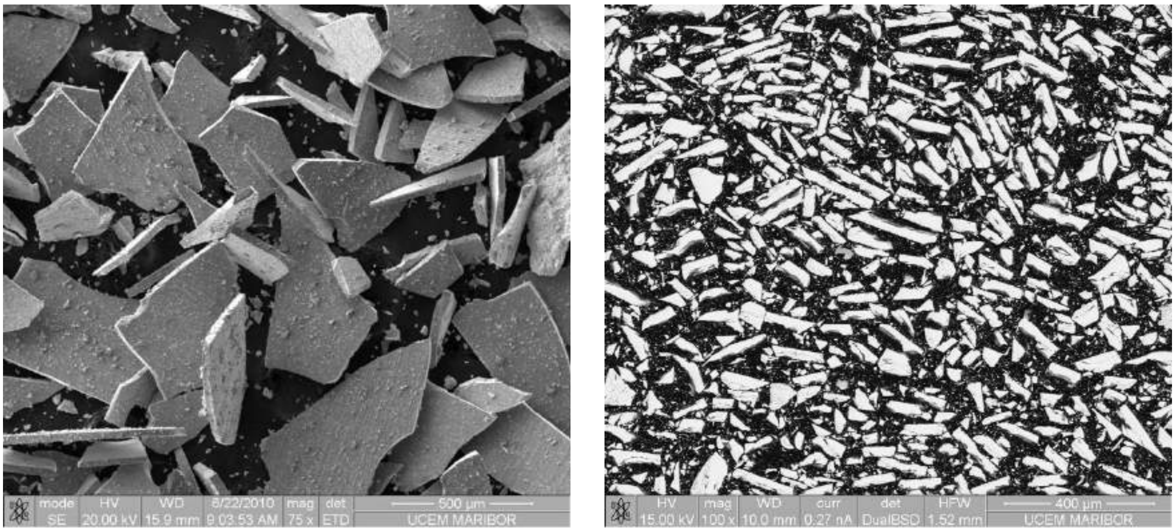
a b

Fig. 1. Co-rotating twin-screw extruder (a) for compounding of polymer bonded magnets and injection moulding machine for net-shape moulding of final magnetic products (b)

The resulting magnetic properties were measured by a permeagraph (Magnet Physik Steingroever). The microstructure of the injection moulded polymer bonded magnets was examined on the transversal cross-section on the metallographic samples with an optical microscope, Nikon Epiphyte 300, equipped with a system for digital quantitative image analysis (Olympus DB12 and software program Analysis). Before metallographic preparation the samples were positioned using metal clamps and carefully cold mounted in epoxy resin. Cold mounting is preferable because of relatively low melting temperature of PPS which could lead to deformation of the samples if using hot mounting procedures. Grinding was performed using SiC papers P320, P500, P1000, P2500, and P4000. Followed by polishing using 1 micron diamond suspension and the final step were polishing using 0, 05 micron colloidal alumina. In both polishing steps a micro-cloth was used which was wetted prior to applying the polishing agent for additional lubrication. Samples were prepared on an automated grinder//polisher, using clockwise rotation 250/40 rpm, and a force of 10N, expect at the final step where the force was reduced to 5N. Additionally, the morphology melts spun NdFeB powder was examined with the scanning electron microscope FEI Siron NC.

3. RESULTS AND DISCUSSION

The magnetic properties of polymer bonded magnets are strongly dependent on size, shape, type, concentration and dispersion of magnetic powder in the polymer matrix. On other hand are the mechanical properties additionally also influence by matrix properties and interfacial adhesion between the magnetic powders and polymer matrix. As long as the size and the distribution of rapidly quenched powders are within the right range, high magnetic properties of polymer bonded magnets can be obtained. This is mainly because the Nd-Fe-B ribbons have high hardness and plate-like shape. The larger the size of rapidly quenched ribbons is, more difficult is to obtain high density of bonded magnets. However, the structure will be destroyed if the size is too small, which results in the deterioration of magnetic properties. It was shown in literature, that the bimodal particle size distribution of the powders enables achievement of high loading factors and lower melts viscosity by injection moulding of bonded magnets [4].

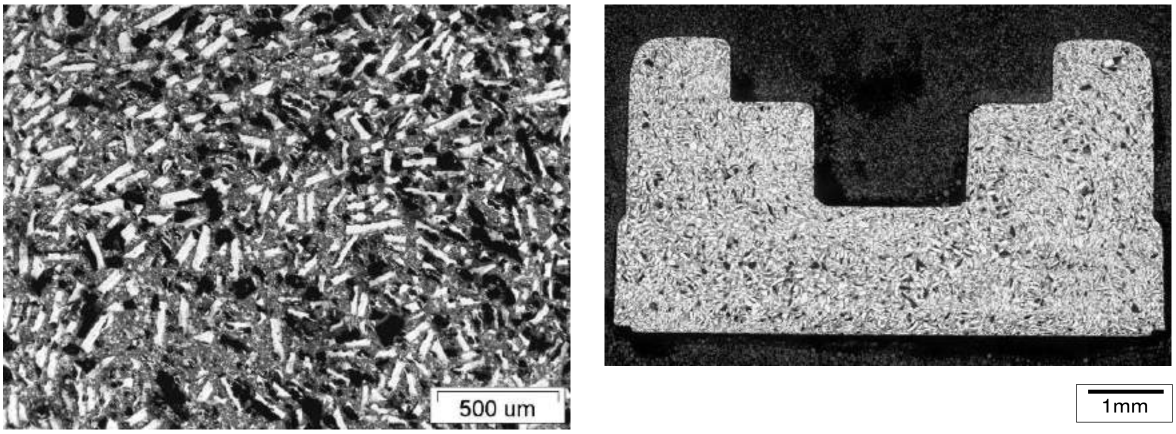


a *b*

Fig. 2. SEM micrographs of plate-like Nd-Fe-B ribbons made by melt spinning (a) and microstructure of polymer bonded magnets (b)

The SEM micrograph on fig. 2. a) shows the morphology of melt spun Nd-Fe-B ribbons. The ribbons are plate-like with a particle size ranging from 10 to 600 μm and average particle size about 250 μm. To achieve higher loading factors and to decrease the melt viscosity during compounding and injection moulding, the melt spun ribbon were grinded and sieved to obtain narrower particle size distribution and average particle size about 150 μm. On the other hand, the SEM micrograph on fig. 2. b) reveals the microstructure of polymer bonded magnet after injection moulding. Its microstructure consists of PPS matrix (shown grey on micrograph on fig. 2.) and homogenously distributes bright plate-like Nd-Fe-B ribbons within the matrix. Additionally, the micrograph on fig. 2. b) reveals also the small amount of porosity in the microstructure of polymer bonded magnets, which is shown as dark regions within the PPS matrix. One would expect the remanence and the energy product of bonded magnets to be directly linked to the amount of binder used. However, porosity and internal magnetic shear loss, also lead to lower the expected B_r values. Pores reduces the magnet density and hence the B_r. They can be limited by particle morphology, type of binder and consolidation technique. Internal shear loss is the effect caused by isolated magnetic particles magnetically shearing with one another within the polymer. The effect increases with higher levels of polymer and powders with low rare-earth content or high B_r/H_{ci} [1].

Based on quantitative analysis of the size and distribution of Nd-Fe-B ribbons in the PPS matrix, we can conclude that the distribution of ribbons in the polymer matrix is bimodal and consisting of larger and smaller ribbons. The average particle size of larger ribbons is from 100 to 150 μm and smaller from 10 to 30 μm, respectively. The micrographs made with optical microscopy at smaller magnifications (fig. 3. a and 3. b) reveal also some degree of microstructure anisotropy which is consequence of plate-like geometry of Nd-Fe-B ribbons and the conditions of melt flow during injection moulding.



a *b*

Fig. 3. Optical micro (a) macrograph (b) of polished cross-section showing the microstructure and orientation of Nd-Fe-B ribbons in the injection moulded polymer bonded magnet

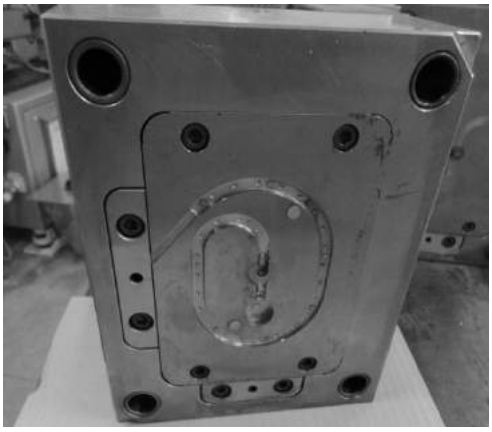
Along magnetic and mechanical properties, the melt viscosity during injection moulding is an important characteristic that describers the porcessability of polymer bonded magnets. To determine the rheological behaviour of the polymer bonded magnets the spiral flow test was used in this research work. For this purpose, a special spiral flow testing mould was designed (fig. 4. a). The spiral flow test is a comparative test for determination of rheological nature of polymers based on measuring the spiral flow length of polymer travelled under specific operation conditions (fig. 4. b).

Table 1. Mechanical and magnetic properties of developed compound for injection moulding of polymer bonded magnets

Properties	Costumers requests	Achieved
Flexure Strength [MPa]	> 70	80,84
Density [g/cm^3]	ca. 5,2	5,16
Br [mT]	0,505 – 0,515	0,509
Hcb[1] [kA/m]	> 340	358,15
Hci [kA/m]	> 800	970,82
$(BH)_{max}$ [kJ/m^3]	> 43	45,40

Finally, after optimizing the volume fraction, average particle size and particle size distribution of the Nd-Fe-B ribbons in PPS matrix, we managed successfully to prepare on the twin-screw extruder granulated compound with desired magnetic and mechanical properties (Table 1.) for injection moulding of rotors onto a steel shaft (fig. 4. c).

1 Coercive Force



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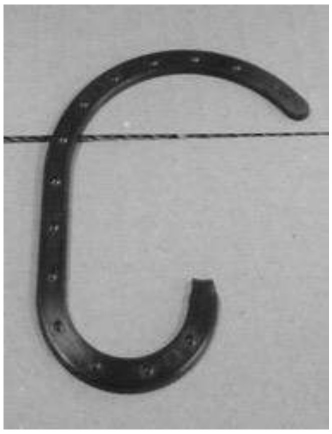

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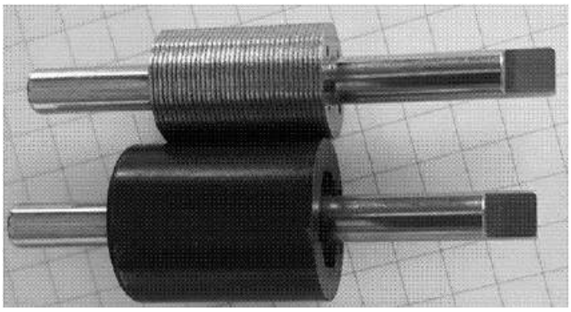

c

Fig. 4. The spiral flow testing mould (a), spiral flow length (b) and injection moulded rotor on a steel shaft (c)

4. CONCLUSIONS

Always shorter life cycles of products and their miniaturization dictate the tempo of development of the new types of polymer bonded magnets. The fight on the global market of polymer bonded magnets is inexorable, the winner will be the manufacturer that is able to produce polymer bonded magnets with the best magnetic (B_r, $(BH)_{max}$, H_{ic}), mechanical and rheological properties, as well as, optimal corrosion resistance. Therefore, the investing in the development of new types of polymer bonded magnets is necessary. The aim of this research work was also to diversify the production programme of the company Magneti Ljubjana d.d. and develop magnetic materials which are tailored according to customers' specific needs, and follow technological trends in high-tech industries, particularly the automobile industry and the manufacturing of sensors, measurement instrumentation and professional electronics. We must be aware that this is a range of customers requiring "zero defects".

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