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## DEPENDENCE OF THE STRUCTURE AND MAGNETIC PROPERTIES OF CAST PLATE-SHAPED Nd<sub>60</sub>Fe<sub>30</sub>Al<sub>10</sub> SAMPLES ON THEIR THICKNESS

### ZALEŻNOŚĆ MIKROSTRUKTURY I WŁAŚCIWOŚCI MAGNETYCZNYCH OD GRUBOŚCI MAGNESÓW Nd<sub>60</sub>Fe<sub>30</sub>Al<sub>10</sub> ODLEWANYCH W POSTACI PŁYTEK

The hard magnetic Nd-Fe-Al alloys are inferior to Nd-Fe-B magnets as far as the magnetic properties are concerned, but their great advantage is that they need no additional annealing to achieve good magnetic properties. These properties depend on the cooling rate from the melting state, and on the thickness of the sample - the best values are achieved at the quenching rates at which the samples have a thickness of 0.3-2 mm. The present study is concerned with the correlation between the magnetic properties of the plate-shaped Nd<sub>60</sub>Fe<sub>30</sub>Al<sub>10</sub> samples and their size - thickness. Two casting ways: with the melt stream perpendicular direction and parallel to the surface of the plates were used. The plates were produced by pressure casting and suction casting. The studies have shown that the cooling rates depends on local propagation on liquid metal in the mold resulting in heterogeneity of structure and properties.

*Keywords:* hard magnetic materials, Nd-Fe-Al alloys, rapid quenching

Stopy Nd-Fe-Al nie posiadają tak wysokich właściwości magnetycznych jak Nd-Fe-B. Ich dużą zaletą jest natomiast możliwość otrzymywania bezpośrednio poprzez odlewanie. Właściwości magnetyczne zależą od szybkości chłodzenia ciekłego stopu, co w praktyce określa się rozmiarem odlewanej próbki. Przy odlewaniu do form metalowych optymalny zakres wymiaru w kierunku odprowadzania ciepła to 0,3-2 mm. W niniejszej pracy zbadano zależność właściwości magnetycznych stopu Nd<sub>60</sub>Fe<sub>30</sub>Al<sub>10</sub> od grubości próbek w postaci płytek w zależności od sposobu odlewania (wtryskiwanie cieczy pod ciśnieniem i odlewanie z zasysaniem) oraz kierunku strumienia cieczy (prostopadle lub równoległe do powierzchni płytki). Badania wykazały, że szybkość chłodzenia zależy lokalnie od sposobu rozchodzenia się cieczy w formie, co skutkuje niejednorodnością mikrostruktury i właściwości.

## 1. Introduction

It is commonly known that the cooling rate strongly effects on the phase structure and even the amorphous phase morphology [1] in rapid quenched materials. Current work is an attempt to find a relationship of cooling rate with the phase structure and magnetic properties on the example of the alloy Nd<sub>60</sub>Fe<sub>30</sub>Al<sub>10</sub>. Even though the Nd-Fe-Al alloys are inferior to Nd-Fe-B magnets as far as the magnetic properties are concerned, their great advantage is that they need no additional annealing to achieve good magnetic properties [2]. Magnetic properties of Nd-Fe-Al alloys depend on the cooling rate from the melting state, and on the thickness of the sample - the best values are achieved at the quenching rates at which the samples have a thickness between 0.3 and 2 mm. Magnets of this size could be used directly, without any further treatment, in electromechanical micro-devices or in the MEMS technique.

In testing of cast amorphous or nanocrystalline materials is often necessary to examine properties of the samples whose sizes vary within a wide range. When using rapid solidification techniques it is difficult to determine the actual cooling rate. Therefore it is important to find a parameter reflecting this behavior. It has been shown that in casting the magnetically hard Nd<sub>60</sub>Fe<sub>30</sub>Al<sub>10</sub> alloy the cooling rate is related to the dimension of the sample measured in the direction of heat removal [3]. In the present study we assumed that the coercivity of the cast Nd-Fe-Al magnets depends on the cooling rate which decides about the microstructure of the material.

It has been found that ribbons subjected to rapid cooling (with the linear velocity of the wheel surface of 30-25 m/s) are amorphous (as examined by X-ray diffraction) and have poor hard magnetic properties, but show certain anisotropy of these properties. Cooling at lower rates gives hardening of magnetic properties [4, 5]. For example the ribbons melt spun at 5 m/s have an amorphous structure

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but also contain some oriented hexagonal precipitates of the Nd phase [4]. Many publications have been devoted to the Nd-Fe-Al materials cast in the bulk form. The authors of ref. [6] examined cylindrical samples of the  $\text{Nd}_{60}\text{Fe}_{30}\text{Al}_{10}$  alloy with diameters ranging from 1 to 15 mm. In another study [7], the authors found that bulk samples with a diameter of 1 mm had the same magnetic properties as ribbon cooled at a rate of 5m/s. According to ref. [8], there is a certain range of the sample size (between 130  $\mu\text{m}$  and 3 mm) within which the samples may be cooled at the rates that ensure producing almost optimum structures, i.e. such that contain the amorphous phase and precipitates of crystalline phases with nanometric sizes.

The literature reports do not however deliver information about the properties of cast Nd-Fe-Al samples which have the form of plates, or about plates cast of other materials such as Nd-Fe-B alloys and the homogeneity of their properties.

In the present study we examined plate-shaped samples cast by various methods. We compared various rapid solidification techniques by analyzing how the coercivity of the various plates depends on their thickness. In casting circular plates with large diameters, a steep temperature gradient is expected to occur during the cooling stage at a certain distance from the inlet of the alloy into the die. In consequence, a gradient of the structural and magnetic properties should appear in the material. This problem was already discussed in our earlier paper [9] in which we reported on casting rods of great diameters. In the present study we examined the structural homogeneity of the plate-shaped samples and its effect on their magnetic properties.

## 2. Experimental

The material examined was the  $\text{Nd}_{60}\text{Fe}_{30}\text{Al}_{10}$  (at%) alloy. The master alloy was prepared from elemental Nd, Fe and Al with a purity of 99.99% or better, by arc melting in a titanium-gattered argon atmosphere. The alloy was melted in a quartz crucible by induction and, then, pushed into a copper die (100 mm in diameter, 50 mm high) under the argon over-pressure in the range of 200-800 mbar. Further in the text this method will be referred as the pressure casting and denoted as PC.

Two variants of the PC process were examined: 1) perpendicular casting in which the molten metal stream is perpendicular to the surface of the plate and hits it at its center, and 2) parallel casting - with the molten metal stream introduced parallel to the surface of the plate (the metal inlet system leading to the cylindrical surface of the plate). The castings had the form of circular plates 22 mm in diameter.

The plates were also produced by suction casting (SC) in which the alloy was melted in an electric arc and then sucked into the die by under-pressure. The plates were rectangular in shape and sized at 10x15 mm.

The die filling degree was determined based on the ratio of the volume of the cast plates to the inner volume of the die, a parameter which represents the coverage of the die volume.

The magnetic properties of the samples were examined using a Lake Shore vibrating sample magnetometer. The samples intended for these examinations were cuboidal

in shape with a 2x2 mm base and the height equal to the thickness of the cast plates. The procedure adopted in selecting the places at which the samples were cut off was as follows. The samples were cut off from at least three different places and their magnetic properties were examined. If there were differences, additional samples were cut off at other places, namely along the sample diameter in both perpendicular and parallel PC plates and additionally along the plate circumference in parallel PC plates. The thermal analysis was performed in a differential scanning calorimeter (DSC) at a heating rate of 40 K/min. The microstructure was examined using a LEO 1530 scanning electron microscope.

## 3. Results and discussion

The  $\text{Nd}_{60}\text{Fe}_{30}\text{Al}_{10}$  alloy has good hard magnetic properties and, what is more, is more suitable than Nd-Fe-B alloys for casting by rapid cooling methods. Fig.1 shows how the die filling degree depends on the thickness of the plate being cast in suction casting (SC) and in casting in which the molten metal is pushed by a gas (PC).

With thin plates, a much better filling of the die was achieved by using the SC technique. With the 0.3 mm plate, the die was filled in 70%, whereas in the perpendicular PC casting the die filling degree was only 50%. In all the techniques examined, the die filling degree increased with increasing thickness of the plate. In the SC process, the 100% filling was achieved for the 0.8 mm thick plate, while in perpendicular PC – for the 1 mm and in parallel PC casting for a plate with a thickness of 1.3 mm.

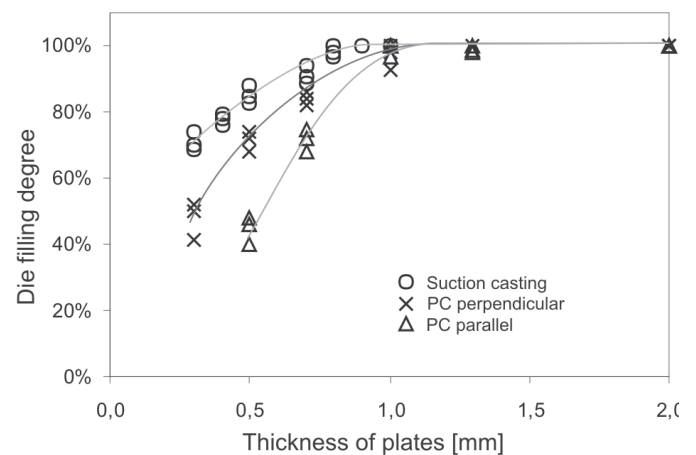


Fig.1. Variation of the die filling degree with the thickness of the plate being cast and the casting technique

Since in the present experiments we were concerned with the plates whose sizes permitted the cooling rate to ensure producing a near-optimum structure [7], i.e. plates with thicknesses ranging from 0.3 to 2 mm (the dimension measured in the direction of heat removal), the magnetic properties of the plates obtained were not significantly different. However, certain differences dependent on the casting method employed were observed (Fig.2).

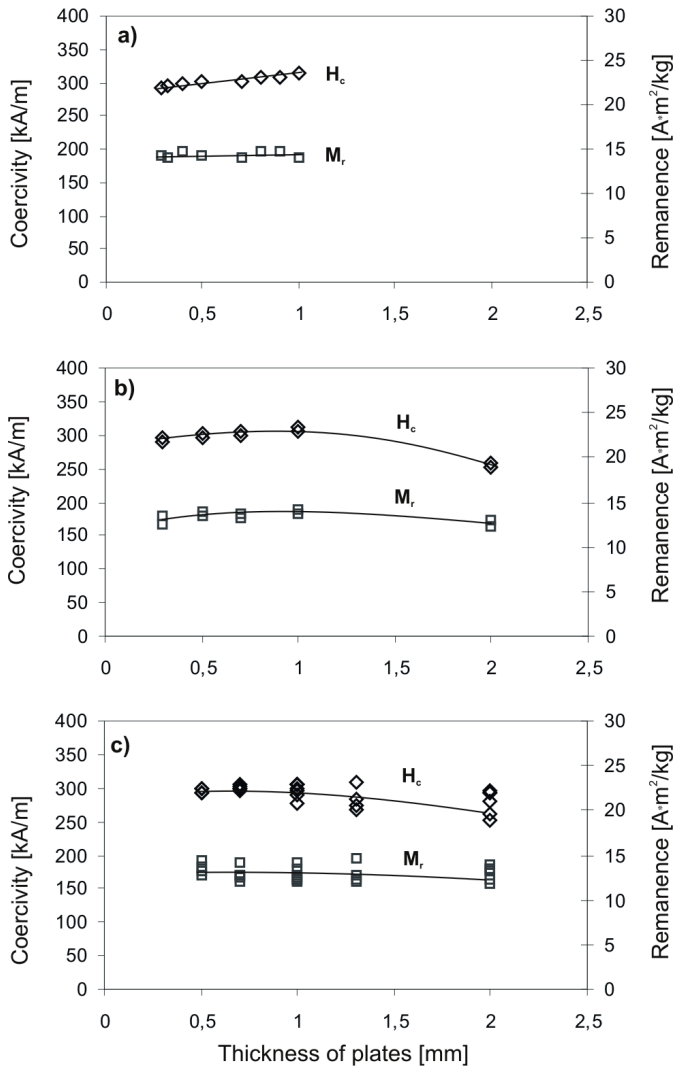


Fig.2. Magnetic properties depending on the thickness of the plates and the casting technique employed: a) suction casting (SC), b) perpendicular pressure casting (PC), and c) parallel PC

In the plates cast by suction casting (Fig.2a), the coercivity slightly increased with increasing thickness of the cast plate: from 293.2 kA/m in a 0.3 mm thick plate to 313 kA/m in a plate 1mm thick. The remanence did not vary and was about  $14.6 \text{ A}^* \text{m}^2/\text{kg}$ . In the plates cast onto the surface perpendicular to the molten alloy stream (Fig.2b) the magnetic properties varied along the plate radius. The highest values of the coercivity and remanence were observed at the sample edges. As the thickness of the plate increased to 1mm, the coercivity and remanence increased, but in the 2 mm thick plate they were lower again.

In figure 2c the magnetic properties of the plates cast by parallel PC were shown. The measured values depend significantly on the measurement point. Unlike in the plates cast perpendicularly, here the magnetic properties do not vary linearly along the plate radius. This may indicate that, during the casting process, the alloy was not cooled uniformly. The thickness of the plate (within the examined thickness range) had no significant effect on the magnetic properties: the coercivity showed a slight tendency to decrease, whereas

the remanence remained unchanged at a value of about  $13.7 \text{ A}^* \text{m}^2/\text{kg}$ .

When comparing the magnetic properties of the plates cast by the various methods one can conclude that the most uniform properties were achieved with suction casting. The magnetic properties of the plates cast by PC were non-uniform and depended on the place where the sample had been cut from the plate. This indicates that the differences in the local cooling rates in the die were greater.

The microstructure of the plates was examined in a scanning electron microscope. The back-scattered electrons images (BSE) of the 1mm thick plates, which were cast perpendicularly by PC are shown in Fig.3. In both examined places the microstructure is varied and separated single needle-like dendrites can be observed. They have length of 4-15  $\mu\text{m}$  and form greater regions when the cooling rate is lower. The peripheries of the samples contained a uniform 'amorphous' phase with precipitates of the equilibrium  $\delta$  phase. Our earlier studies have shown that this 'amorphous' phase is composed of Fe and Nd rich nanocrystallites (5-30 nm), embedded in an amorphous matrix [8] (for the simplicity, it will be referred to as the 'amorphous' phase). With increasing distance from the sample edge the share of this amorphous phase decreases, whereas the amount and size of the  $\delta$  precipitates significantly increases.

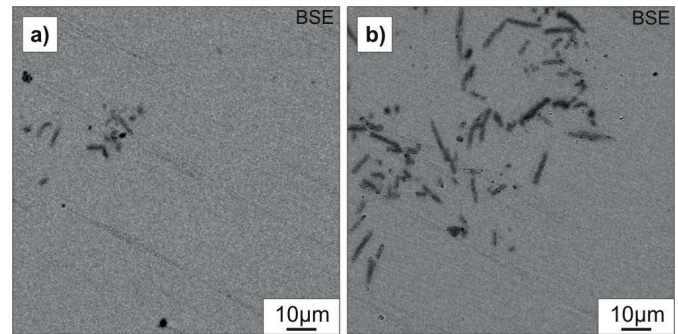


Fig.3. BSE images of the surface of the 1 mm thick plates cast by perpendicular PC: a) region near the sample edge, b) central region of the sample

Because of the existence of the structural gradient, the measured value of the coercivity depends on the area from which the sample is cut out from the cast plate. It was shown that the magnetic properties of a circular plate vary along its radius. For example in a 1mm plate, the coercivity was 311 kA/m at peripheries while it decreases to 305 kA/m in the central region.

Figure 4 shows images of the 1 mm thick plate cast by parallel PC. Furrows on the plate surface indicate that the die was filled in a stepwise way (Fig.4a, b). These furrows suggest that the injected alloy was rebounded from the bottom of the die and filled its side regions moving up to its top. The central and upper parts of the cast formed during the final stage when the die was already hot. Local changes of the microstructure are related to variation of the cooling rate respectively to the sequence of the die filling. The regions that formed first have a homogeneous structure and were composed of the 'amorphous' phase (Fig.4c), whereas the regions where the die was filled later contain additionally dendritic precipitates

of the equilibrium  $\delta$  phase (Fig.4d). The precipitates were the most numerous in the upper part of the plate.

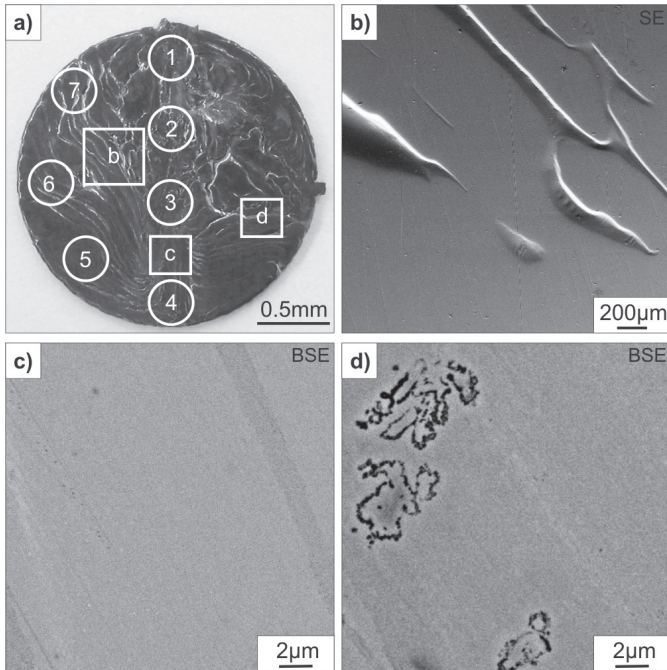


Fig.4. Images of the surface of a 1 mm plate cast by parallel PC: a) entire surface of the plate with an indication of the regions of microscopic observations and magnetic measurements, b) topography of the furrows observed on the sample surface (secondary electrons SE), c) and d) BSE images of the surface areas indicated in the photograph (a)

In the plates cast by parallel PC, no unequivocal relation between the magnetic properties of the sample and its radius can be observed. The regions, for which the magnetic parameters are given in TABLE 1 were indicated in Fig. 4a. In these plates (parallel PC), the coercivity and remanence vary along the plate diameter, with the highest values being observed at the plate bottom. Comparing the properties of the samples when passing from those cut out from the upper regions of the plate to those cut from its side regions one can observe that the magnetic properties become worse. This can be attributed to the cooling rate being slower there.

TABLE 1  
Magnetic properties of a 1mm plate cast by parallel PC  
(measurement points are indicated in Fig.4a)

	1	2	3	4	5	6	7
Hc [kA/m]	278.0	294.6	304.9	305.0	304.92	300.6	289.4
Mr [A*m <sup>2</sup> /kg]	10.79	12.32	12.69	12.56	12.76	12.28	11.39

In figure 5 the DSC curves obtained for samples cut from the 1 mm plate cast by parallel PC were shown. Micro-calorimetric examinations revealed differences in the characteristic temperatures. The temperature of the peak

of the exothermal effect in sample 1 is  $T_p=548^\circ\text{C}$  whereas in samples 2, 3 and 4 it is  $555^\circ\text{C}$ . This indicates that there are local differences in the chemical composition of the amorphous phase.

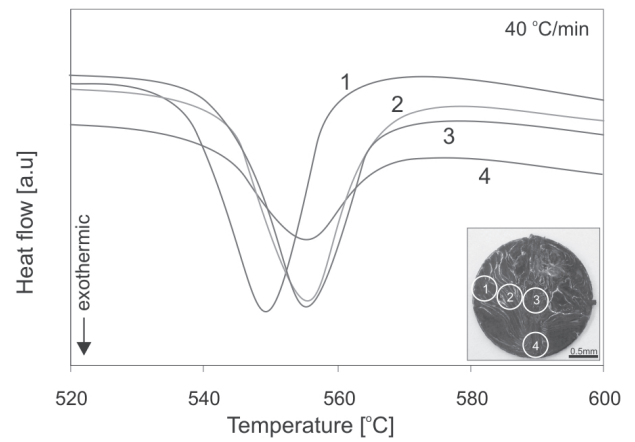


Fig.5. DSC curves obtained for the samples cut out from a 1 mm thick plate cast by parallel PC (the inset indicates the regions from which the sample was cut out)

#### 4. Summary

$\text{Nd}_{60}\text{Fe}_{30}\text{Al}_{10}$  alloy plates produced by suction casting (SC) and pressure casting (PC) were examined. The PC processes were conducted in two versions: perpendicularly and parallelly to the plate surface. It has been found that for thin plates a much better filling of the die was achieved by using the SC technique. Moreover, in this case, the magnetic properties were distributed more uniformly. The plates cast by perpendicular PC were characterized by a gradient of the structure and magnetic properties along the sample radius. The best magnetic properties were observed at the sample edges, which indicates that the central part of the sample was cooled at a lowest rate and, in consequence, great amounts of the equilibrium  $\delta$  phase was formed there. A similar gradient of the structure and properties was observed in the plates cast by parallel PC. However the gradient formed not along the sample radius but according to the sequence in which the individual regions of the die were filled during the casting process. The plates cast by this method showed the greatest differences in their magnetic properties.

Using the SC casting method it is possible to produce 10x15 mm plates homogeneous in their structure and magnetic properties. The plates cast by the other methods were greater, but even in the region with the size comparable with the size of the SC plates, a gradient of the structure and, in consequence, of magnetic properties appeared.

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