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Magnetic induction of polymer composites filled with ferrite powders

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ABSTRACT

Purpose: The goal of this work was to determine influence of amount and type of ferrite fillers on magnetic induction of epoxy composites. Six mixtures that contain different amount of ferrite powder were prepared. Additionally an effect of type and amount of introduced ferrite powder on epoxy resin processing conditions and parameters was searched.

Design/methodology/approach: In this research the method of preparing polymeric gradient composites was centrifugal casting. The experimental procedure focused on evaluating the magnetic induction of gradient composites. Magnetic induction of composites based on epoxy resin, which contain ferrite powders was measured using milliteslometer. Measurements were performed for three filler contents and for two types of filler.

Findings: Centrifugal casting method allowed obtaining materials with different percentage of ferrite content in subsequent layers of cylindrical composites. Moreover it was observed that values of magnetic induction were higher for composite with barium ferrite than for composite with strontium ferrite but the difference was not too high.

Research limitations/implications: The main problem of this work was about limitation of maximum level of fillers content. Adding more filler than 30%vol caused very rapid composite viscosity increase and made air removing and casting impossible. For that reason there were made composites with 10%vol., 20%vol. and 30%vol. of ferrite powders. Trials were performed with mixtures that contained up to 50%vol. of filler.

Practical implications: Method applied in this research allowed to obtain materials that are characterized by gradient of magnetic properties. Such composites find applications in electrotechnical industry and in mechanical engineering.

Originality/value: New polymeric gradient materials were developed using centrifugal casting technology. Magnetic properties of these composites were determined depending on distance from the surface of the test piece.

Keywords: Polymers; Magnetic properties; Casting

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MATERIALS

1. Introduction

Composite materials are one of the main types of the engineering materials, next to metals and alloys, ceramics and polymeric materials. They are made at least of two separate types of substances each with its own characteristics, one of them is called matrix while the second is filler. In composite materials these two phases are immiscible and are separated by boundary interface layer. Such a wide range of constituent materials gives materials scientists very big field of possibilities to create new composites and new generations of composites. By proper innovative material selection and design it is possible to change properties of products depending on requirements and future applications. Composite materials find practical application in many domains of industry for instance in civil engineering, machine building, sport and leisure industry, automotive industry, aerospace industry etc.

Nowadays some kinds of composite materials demonstrate gradual distribution of structure and/or composition, e.g. gradual distribution of fillers in the matrix. Such continuous structure or composition changes can eliminate one of basic drawbacks of other composites, namely sharp boundaries between joined substances and stresses concentrations at the same time. In this way scientists try to increase strength properties of ready products and to differentiate properties across them. These, just presented materials from technical point of view are named Functional Graded Materials (FGMs). For the first time they had been manufactured in 1944 but until the end of 20th century they were not interesting for science and industry. Intensive researches on these materials began approximately from 1987 and wide knowledge about these materials successfully was developed [1,2].

A special class of gradient materials are polymeric FGMs. The matrix in polymer graded materials usually is made of thermosetting or thermoplastic polymers. Until now scientific researches have comprised and in papers have described many different combinations of materials applied to form graded composites. Among them from scientific and industrial point of view the following material configurations are very interesting: PMMA/glass fibre [3], epoxy/carbon fibre [4], SiC/epoxy [5], PVC/PMMA [6], epoxy/graphite [7, 8], epoxy/strontium ferrite [9] etc. Also there are very numerous methods and manners of PGMs manufacturing.

Magnetic polymer materials which are characterized by specific distribution of filler, allow to obtain gradual changes of magnetic properties throughout the specimen’s cross-section. These materials up till now have been examined rather rarely. For this reason in the present study authors decided to search magnetic properties of designed epoxy composites, which were produced by rotational casting.

The space surrounding a magnet or a guide with flowing current is called the magnetic field. It is the space in which there are magnetic forces acting mechanically on permanent magnets and wires with electric current and causing the induction of electric current in a wire moving in this space. The SI unit of magnetic field intensity is called the tesla [T] but in CGS this unit is named gauss (1G = 10^{-4} T) [10-12]. Physical cause of magnetic fields generation is the movement of electrons inside atoms or molecules of permanent magnets, or in the conductor in the form of electrical current. The magnetic field presence is manifested as a system of forces acting along magnetic forces lines. Magnetic forces lines are curves tangent to magnetic forces course at every point. Magnetic forces direction is determined by the direction of magnetic field [11, 13]. When in the magnetic field produced by electric current, lines of magnetic forces are closed, what means they are without beginning or end, it is said that field is strong and the magnitude of magnetic induction is large (close to the conductor). On the other hand where these magnetic forces lines are opened (apart from conductor), the field is weak and the magnitude of magnetic induction is small [14, 15].

Depending on the intensity of the magnetic field needed to achieve magnetization of given material and the durability of magnetization after removing the external magnetic field, materials are divided into [14-18]:

- hard magnetic materials (magnetization in strong magnetic fields with intensity higher than 10^3 A/m; magnetization is durable – it disappears under the influence of the opposite strong magnetic field): they are used for making permanent magnets that can for a long time maintain strong magnetic field,
- soft magnetic materials (magnetization in the weak magnetic fields with intensity lower than 10^3 A/m; undurable magnetization which disappear almost instantly after removal of external magnetic field): they are used for cores of transformers, generators and motors.

Furthermore, taking into account phenomena in which magnetic materials are involved, they are divided into 5 categories [10-12, 15, 16, 19, 20]:

- *ferromagnetics* - magnetizes very strongly in the direction consistent with that of the external magnetic field. Ferromagnetics retain magnetization after disappearance of the external magnetic field. Each ferromagnetic material was found to have a definite temperature, the Curie temperature, above which ferromagnetism disappears and the material becomes only paramagnetic (for instance Curie temperature of iron is 770°C). Examples of ferromagnetic materials are: iron, cobalt, gadolinium and nickel,
- *paramagnetic materials* - magnetize in a very low degree, but in direction that also is consistent with the direction of the external magnetic field. Paramagnetics include: tungsten, platinum, magnesium, tin, aluminium
- *diamagnetic materials* - magnetize in a very low degree in the opposite direction to the direction of the external magnetic field. Diamagnetics abate action of magnetic field. Magnetization is proportional to the intensity of the external field and is independent of temperature. Diamagnetic phenomenon is induced by electromagnetic induction and it is independent of atomic structure and his properties. For this reason diamagnetism is observed in all substances that are located in magnetic field. Examples of this materials are the followinh: water, gold, coal, copper, silver, neon, helium, argon, bismuth etc.,
- *ferrimagnetic materials* - depending on the type of dipoles permanent alignment, two more types of magnetic materials are distinguished. In ferrimagnetic materials dipoles are antiparallel. The main ferrimagnetics are magnetic oxides called ferrites.
- *antiferromagnetic materials* - in these materials dipoles are paired off in opposite directions. Antiferromagnetic materials include manganese dioxide, chromium, some rare earth metals and compounds in the composition, which include at least one element of a group of transition metals. Chromium is the material in which quantum mechanical effects make the spins alternate. Antiferromagnetic materials have a *Néel temperature* T_N (the Néel temperature is named after Louis

Néel) above which the materials become paramagnetic and the spins suddenly become random.

Ferromagnetic, ferrimagnetic and antiferromagnetic materials are strongly magnetic (up to the definite temperatures interact intensely with a magnetic field), whereas paramagnetic and diamagnetic materials are weakly magnetic (at any temperature these substances interact weakly with a magnetic field) [10].

Interaction of atoms contained in the crystal grid nodes can overcome destructive heat flow and cause the orientation in defined order of magnetic moments (dipoles). This arrangement is in general parallel alignment of adjoining dipoles (ferromagnetism), or antiparallel (neighbouring carrier is parallel but contrary direct - antiferromagnetism - neighbouring moments have the same value). Ferrimagnetism is when neighbouring moments have different values [20].

Moreover Hurd distinguishes even ten types of magnetic ordering: metamagnetism, ferromagnetism incipient, superparamagnetism, asperomagnetism, helimagnetism ideal spin glass and cluster glass, mictomagnetism and sperimagnetism, etc [21, 22].

In polymeric composites fillers with strong magnetic properties are used. All problems discussed up to this moment are also actual for polymeric materials. They are far more complicated because of matrix presence and because of different interactions between magnetic particles and interactions between particles and matrix. Also the process of composite formation is very important. In many cases magnetization is performed during composites formation and matrix curing.

Even more complex phenomena take place when gradient composites are considered. Also properties of gradient magnetic composites are very hard to describe and to foresee. It is very little said about these problems in scientific literature. Presented research is experimental in his charactere.

2. Materials and methods of research

In research programme the following materials were used: epoxy resin (Epidian 6) and curing agent (triethylenetetramine) obtained from Nowa Organika-Sarzyna (Poland) that were used as a matrix. While as fillers two types of ferrite powders (Fig. 1) were used: barium ferrite ($BaFe_{12}O_{19}$) and strontium ferrite ($SrFe_{12}O_{19}$). Both types of powders were obtained from ZAM Trzebinia (Poland). Chosen characteristics of used materials are presented in Table 1 to Table 4.

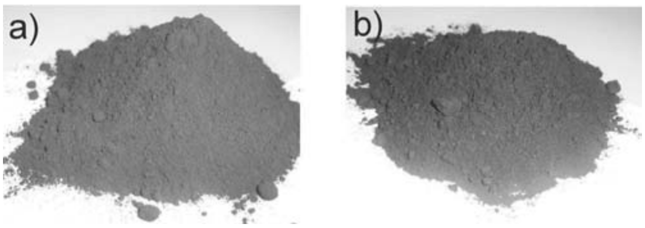

Fig. 1. Powders: a) $BaFe_{12}O_{19}$,b) $SrFe_{12}O_{19}$

Table 1.
Main characteristics of epoxy resin Epidian 6

Density (20ºC)	1,17g/cm³
Viscosity (25ºC)	10000-15000 mPa·s
Boiling point	> 200ºC
Melting point	-
Ignition temperature	> 200ºC
Autoignition point	> 300ºC

Table 2.
Specification of curing agent - triethylenetetramine (TETA)

Trade name	Z-1
Form	colourless liquid
Density (25ºC) [g/cm³]	0,98
Viscosity (25ºC) [mPa·s]	20-30
Amine value [mg KOH/g]	min. 1100

Table 3.
Main characteristics of barium ferrite powder

Chemical formula		$BaFe_{12}O_{19}$
Form		powder
Fe_2O_3 [mol] BaO [mol]		5.6-6.2
Humidity % max		0.5
Density (20ºC) [g/cm³]		5.3
Melting point [ºC]		1315.6
Formula weight [g/mol]		1111.46
Solubility		water-insoluble
Smell		inodorous
Grain size [µm]		<100
Component %	Fe	58.6-59.6
	Ba	12.7-13.7
	Mn max	0.5
	$BaSO_4$ max	1.0
	SiO_2	0.3-0.6

Table 4.
Main characteristic of strontium ferrite powder

Chemical formula		$SrFe_{12}O_{19}$
Form		powder
Fe_2O_3 [mol] SrO [mol]		5.6-6.2
Humidity % max		0.5
Density (20ºC) [g/cm³]		4.9±0.2
Melting point [ºC]		-
Formula weight [g/mol]		1061.77
Solubility		water-insoluble
Smell		inodorous
Grain size [µm]		<100
Component %	Fe	61.4-62.4
	Sr	8.6-9.6
	Mn max	0.5
	$SrSO_4$ max	1.0
	SiO_2	0.3-0.6

In order to produce specimens one of the most effective method for preparing polymer gradient materials (PGMs) was selected, namely it was rotational casting. Also in the literature it is described as popular method that could be successfully used for manufacturing of FGMs. Technological details of this method depend among others on the type of formed material. Three main forms are applied: liquids, pastes or powders. For the purpose of this research it is important that using this method of manufacturing it is possible to control on the gradient properties for example by altering rate and time of centrifugal casting but, in the case of filled composites, also by size, shape and amount of particles [23–26].

In the first stage of composition preparing all weights of components were calculated in order to achieve the following volume fractures of fillers 10%vol., 30%vol. and 50%vol. First the filler's particles were mixed with epoxy resin. Then curing agent was added to this mixture and once more all components were mixed together. In the next step mixtures were left for a moment in order to allow the mixture to deaerate. Without deaeration many defects like voids and bubbles were created in ready composites. As a result of trials of formulations preparing and casting it came obvious that applying higher fillers concentrations than 30%vol. causes very significant increase of viscosity which makes it impossible to achieve good samples with centrifugal casting of prepared composites. For this reason, even that it was possible to introduce higher amounts of filler into the matrix, compositions with 10%vol., 20%vol. or 30%vol. of powders were used in the next stage of the research. In future studies authors consider other methods of filler introduction into the matrix and other filler-matrix systems in order to achieve higher concentrations.

In the next step prepared mixtures were cast into the font mould fixed in rotational device and then rotations were switched on (Fig. 2). Mould with processed mixture was rotated around one horizontal axis. Rotational speed was then increased continuously up to the final one. That mixture was centrifugally cast with a rotation speed of 446 rpm. This speed was chosen to overcome gravity force during casting. All specimens were produced in the same conditions and using the same parameters. This procedure was repeated for each mixture. During the rotational casting process the mixture was hardened.

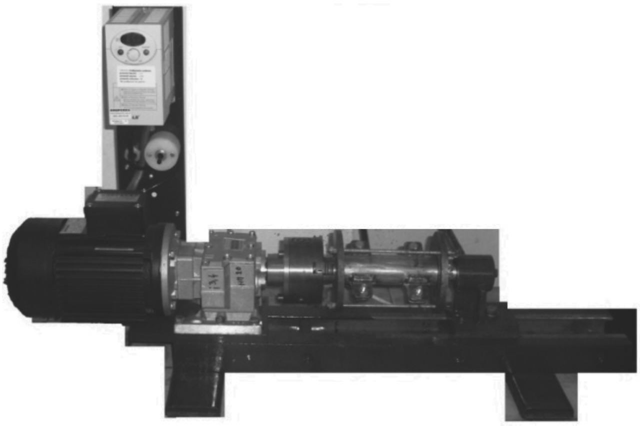

Fig. 2. Rotational casting device with inverter

Higher value of density of ferrite powders than matrix density and action of centrifugal force evoked sedimentation in composites during the rotational casting. Filler powders migrated and settled down gradually on the form's walls as a result of applied centrifugal force. Finally this centrifugal force acting in radial direction of mould caused that almost all particles had been accumulated near the outer mould's wall.

It is possible to change the migration velocity of solid particles by rotational speed regulation and by resin viscosity changes. It is being planned in future research works. In the present research fillers concentration and the type of fillers were changed.

3. Results and discussion

Applied rotational casting technology and the sedimentation phenomenon allowed to produce polymer composites with various filler content throughout the thickness of specimens. Gradient of filler concentration was achieved and in the same way gradient of properties. Magnetic induction was measured with hallotronic measuring device Elimag 2 (Czech Republic - Fig. 3).

Fig. 3. Hallotronic measuring device Elimag 2

Three measuring directions (a,b,c) and measuring points are presented in Fig. 4. Results of magnetic induction for epoxy-barium ferrite and epoxy-strontium ferrite composites as a function of radial distance from interior surface of samples are shown in Fig. 5-7. Curves for two specimens with 10%vol. of two types of ferrite powders are presented (Fig. 5). Measurements were made on the cross-sections of specimens (Fig. 4-a). As can be seen the highest result of magnetic induction obtained for composite with strontium ferrite was (1,6 mT). Whereas the highest result for composite with barium ferrite achieves the value 1,4 mT. The highest value of magnetic induction for both specimens with 20%vol. of ferrite powders obtained circa 7 mT (Fig. 6), meanwhile for specimens with 30%vol. of fillers - 9.3 mT ($BaFe_{12}O_{19}$) and 8.8 mT for $SrFe_{12}O_{19}$ (Fig. 7). Additionally as can be seen in most experimental points composite with barium ferrite obtained higher values of magnetic induction than composites with strontium ferrite.

Fig. 4. Measurements directions and points on specimen

Dependence of magnetic induction values as a function of axial distance from the beginning of the test piece (Fig. 4-b) is presented in Fig. 8-10. Measurements were made on exterior surface of ready composite samples and begun from edge of specimen. Applied 10%vol. fracture of barium ferrite in composites allowed to receive magnetic induction on 2.6 mT level, while for the other composite obtained induction was 5.9 mT (Fig. 8). The highest value of magnetic induction for both specimens with 20%vol. of ferrite powders obtained over 9 mT (Fig. 9), meanwhile for specimens with 30%vol. of fillers - 11.3 mT ($BaFe_{12}O_{19}$) and 10.4 mT for $SrFe_{12}O_{19}$ (Fig. 10). It can be also observed that for some distances (in the range 3-16 mm) results for both ferrite composites were very similar. The course of these curves is typical for all magnets. The highest induction is at edges (poles) and the smallest is in the middle of the magnet.

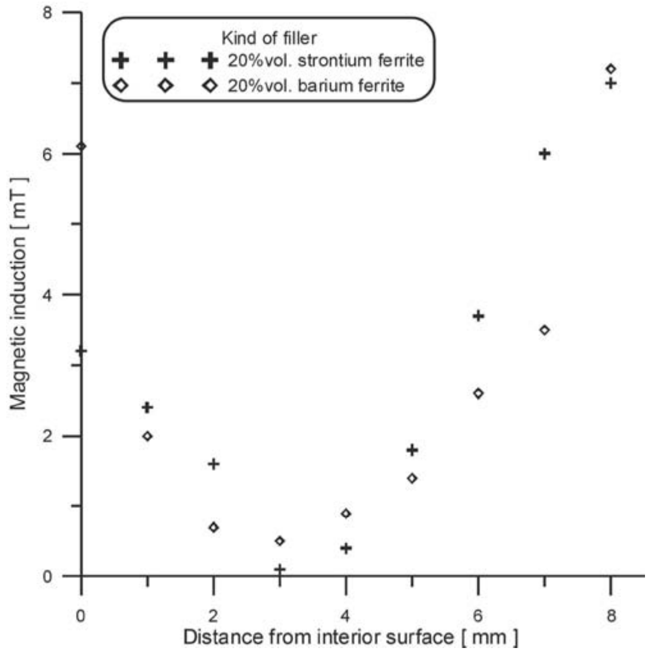

Fig. 6. Results of magnetic induction that where measured on cross-section of specimens for 20%vol. of fillers

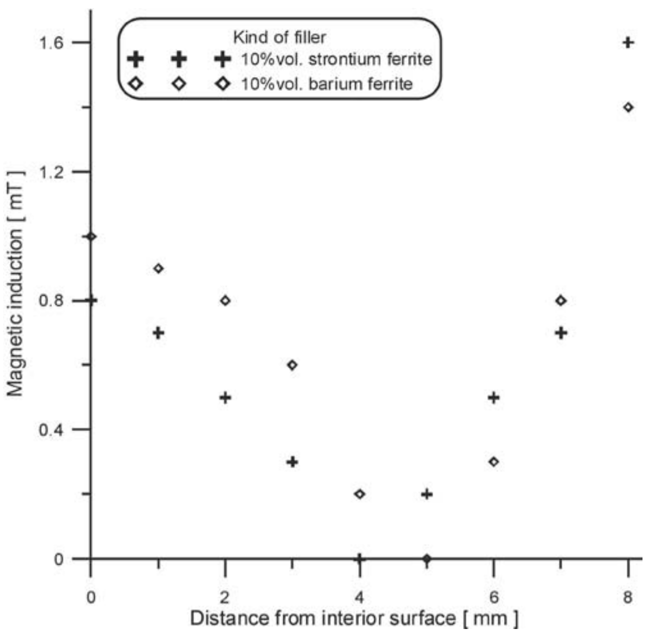

Fig. 5. Results of magnetic induction that were measured on cross-section of specimens for 10%vol. of fillers

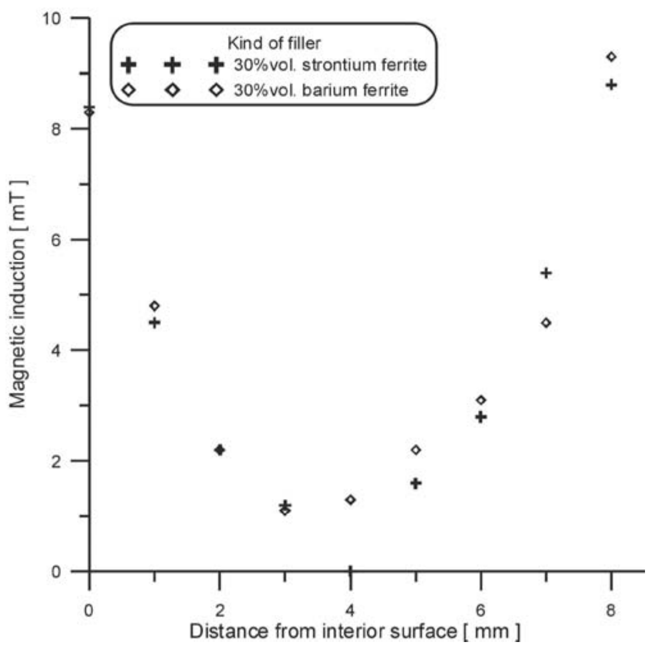

Fig. 7. Results of magnetic induction that were measured on cross-section of specimens for 30%vol. of fillers

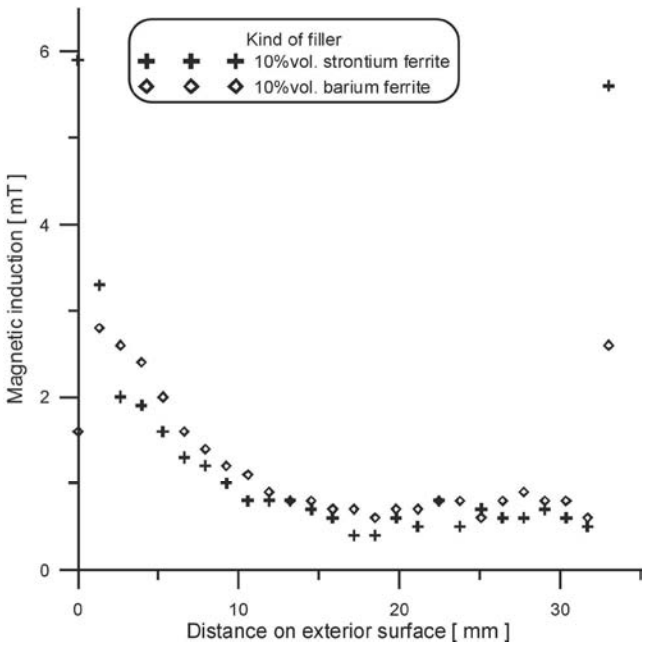

Fig. 8. Magnetic induction values for 10%vol. of fillers on exterior surface of the test piece

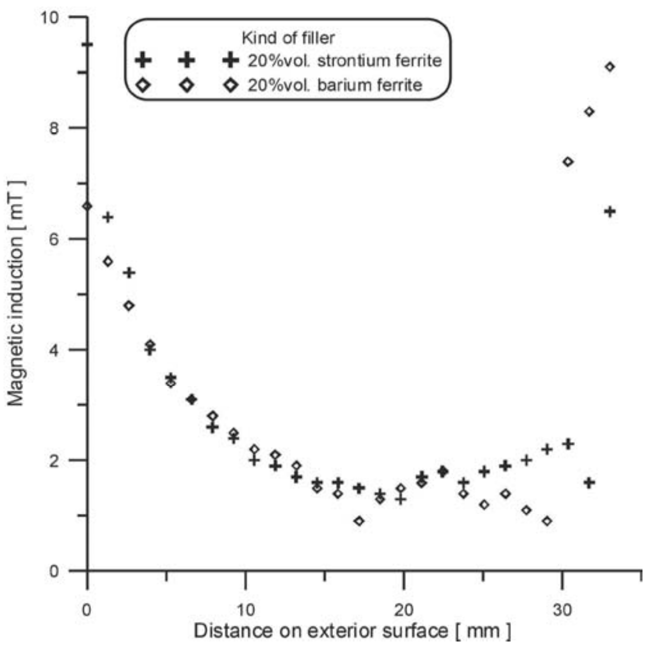

Fig. 9. Magnetic induction values for 20%vol. of fillers on exterior surface of the test piece

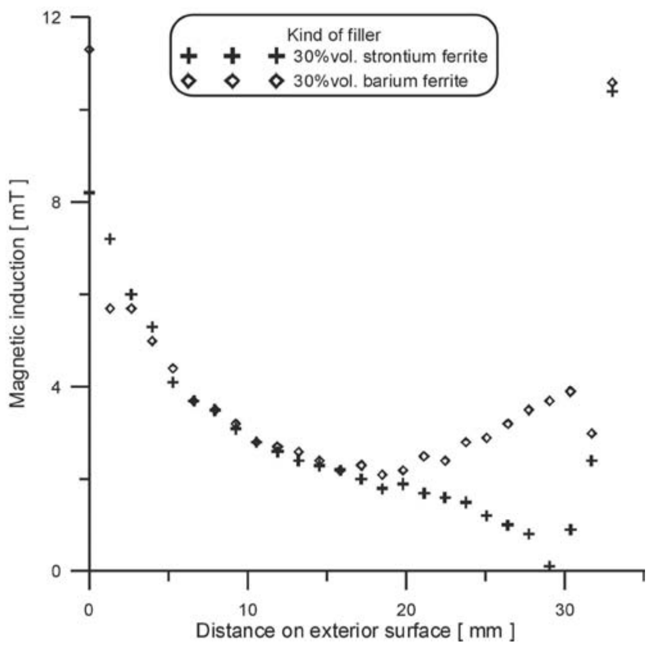

Fig. 10. Magnetic induction values for 30%vol. of fillers on exterior surface of the test piece

Differences between external and internal results were caused by distribution of ferrites in composites what was obtained by using centrifugal force applied during the process of manufacturing.

In Fig. 11 there are presented curves for three contents of strontium ferrite: 10%vol., 20%vol. and 30%vol. As it could be expected higher magnetic induction values where obtained for higher amounts of filler in composite materials. Similar chart (Fig. 12) was made for every filler contents in epoxy-barium ferrite composites.

The measurements were made on outer surface of composites. For the lowest content of ferrite (10%vol. of filler) the highest value of magnetic induction was 5,9 mT for strontium ferrite and 2,6 mT for barium ferrite. While for 20%vol. of filler magnetic induction was 9,5 mT for strontium ferrite composite and 9,1 mT for barium ferrite composite.

The last chart (Fig. 13) presents correlation between magnetic induction and circumferential position on specimen with 10%vol.of strontium ferrite (Fig. 4-c). As it can be observed results are contained in narrow range of magnetic induction values (0.4-1.2 mT). Thus, no essential differences between measurements results were observed on circumference of test pieces. It is in good agreement with expectations. No essential differences in filler content on circumferential position were observed.

Achieved results indicate that applied rotational casting method allows to produce polymer composites that exhibit significant magnetic properties and allows to form significant gradient of magnetic properties in radial position.

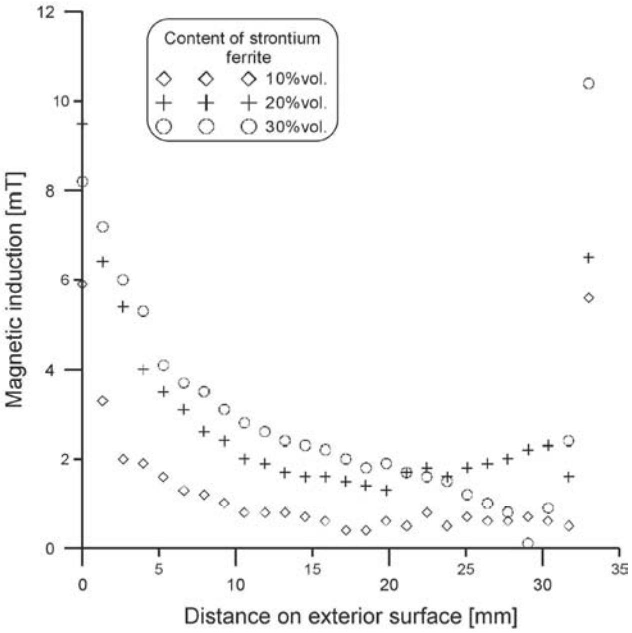

Fig. 11. Results for composites with strontium ferrite that where measure on exterior surface

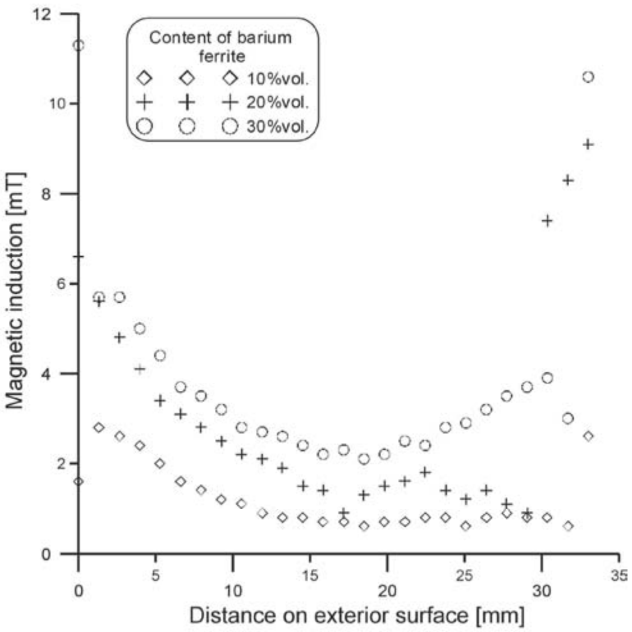

Fig. 12. Results for composites with barium ferrite that where measure on exterior surface

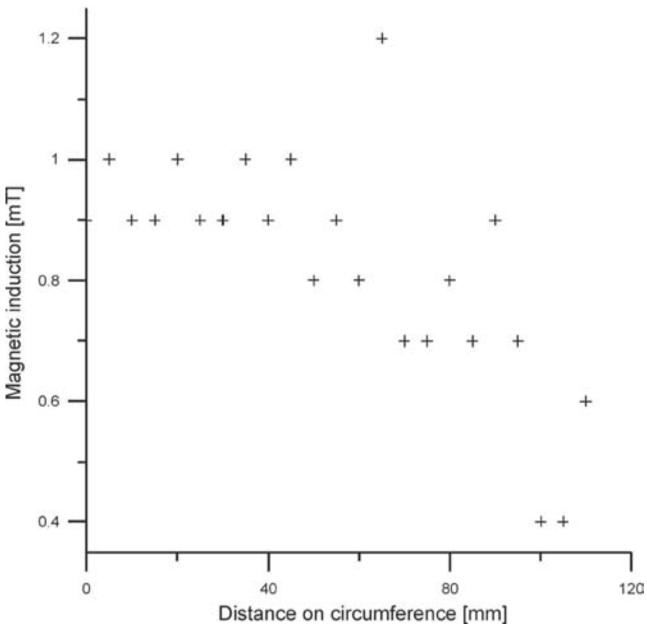

Fig. 13. Relation between magnetic induction and distance on circumference of specimen with 10%vol.of strontium ferrite

4. Conclusions

This study was carried out in order to determine the magnetic induction values of the polymeric gradient composites. Basing on the obtained results the following conclusions were drawn:

- It is possible to produce polymer composites that exhibit significant magnetic properties.
- By using centrifugal casting it is possible to prepare compositions that contain up to 30%vol. of ferrite powders. Higher amount of fillers increased viscosity, which did not allowed to prepare good samples.
- Maximum values of magnetic induction in radial position were obtained near exterior surface of the sample (1,4mT for 10%vol. of barium ferrite, 1,6mT for 10%vol. of strontium ferrite).
- Maximum results of magnetic induction were obtained for 30%vol. of barium ferrite 11,3mT while for strontium ferrite it was 10,4mT.
- Comparison between two types of ferrite powders showed that the results for both had been comparable.
- In future research authors want to increase content of filler by viscosity modification of mixture matrix for instance by introducing thinner.

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