

Use of the Graphite to Obtain Composites for Absorbing Electromagnetic Radiation

Denis Miroschnichenko¹, Vladimir Lebedev¹, Maksym Riabchenko¹, Ruslan Kryvobok¹, Anna Cherkashina¹, Georgiy Lisachuk¹, Yevhen Soloviev¹, Petro Stukhlyak², Andriy Mykityshyn²

¹ *National Technical University «Kharkiv Polytechnic Institute», 61002, Kirpychova str.2, Kharkiv, Ukraine*

² *Ternopil Ivan Puluj National Technical University, Ruska str. 56, Ternopil, Ukraine*

Received February 21, 2023; Accepted July 3, 2023

Abstract

The aim of the article is to study graphite for the production of polymer ceramic-inorganic compounds with desired properties. The technological characteristics of polymer ceramic-inorganic composites based on polyamide, modified with ceramic-inorganic graphite-ferromagnetic fillers: silicon carbide, chromium oxide Cr_2O_3 and graphite, were determined. It is shown that binary modification with ceramic-inorganic graphite-ferromagnetic fillers of composites based on polyamide 6 - SiC 10% wt - Cr_2O_3 10% wt. and polyamide 6 - SiC 10% wt. - graphite 10% wt. systems is optimal for technological characteristics improvement. For three-component complex modification with ceramic-inorganic graphite-ferromagnetic fillers, the most optimal system is polyamide 6 - SiC 10% wt. - Cr_2O_3 10% wt. - graphite 10% wt. It was found that received polymer ceramic-inorganic composites for absorbing electromagnetic radiation can be processed by high-performance injection molding and extrusion methods according to their technological characteristics.

Keywords: *Polymer; Ceramic-inorganic; Composites; Fillers; Absorption; Electromagnetic radiation, Technological characteristics.*

1. Introduction

An important direction in modern materials science are studying materials for electromagnetic radiation absorption. Their wide application is mostly associated with information technologies rapid development that caused modern electronic pollution appearance, such as electromagnetic interference and radiation, electrical noise, radio frequency [1]. That is why the need for electromagnetic shielding is increasing every day in order to avoid unwanted electromagnetic waves interference from electromagnetic pollution and to protect against it [2]. Today, the new materials design for electromagnetic radiation absorption with small thickness, low density, transmission wide range and absorption is underway. They can help in solving problems with radio absorption of electromagnetic radiation. Receiving such materials with high technological, operational and economic characteristics is very perspective.

One of the promising components of substances that have a high absorbing ability in relation to electromagnetic waves is graphite. Graphite, in turn, is obtained by the graphitization of coal tar, and coal tar must be characterized by a certain set of physicochemical parameters [3-5].

Looking to technological, operational and economic criteria the polymer composites for absorbing electromagnetic radiation based on thermoplastic matrices with inorganic fillers addition, such as polymer ceramic-inorganic composites, are the most perspective [6-13]. Such materials not only combine components' properties but also acquire new properties that are not inherent to individual components. At the same time, during products and parts design for electromagnetic radiation absorption from composite polymer materials, the attention fo-

cus, along with their structural features, is also always the material technological characteristics. technological characteristics must be processed using certain technologies and ensuring the specified products and parts parameters. While products and parts design for electromagnetic radiation radio absorption from polymer ceramic-inorganic composites is parallel decision-making both about the materials composition used and about products design parameters, and at the same time it is necessary to take into account manufacturing technologies. So, it is necessary to create an integrated cycle that includes both separate systems for parts design and production for electromagnetic radiation radio absorption from polymeric ceramic-inorganic composites, as well as an up-to-date materials database. Thus, the parts and product design for electromagnetic radiation radio absorption from polymer ceramic-inorganic composites is a very difficult task especially when we have to create new materials. It comes because it requires appropriate technological and calculation information, for the products themselves, the equipment for their manufacture, as well as various technical parameters of their processing. At the same time, the products and parts' high quality and durability for electromagnetic radiation radio absorption from polymer ceramic-inorganic composites is due to the next complex: correct material choice with appropriate technological characteristics and selection of the most effective method of their processing.

In modern scientific sources, there are a lot of research on receiving materials for electromagnetic radiation radio absorption. Most research concerns the effective materials design for electromagnetic radiation radio absorption based on mixtures and composites of fillers with magnetic properties (such as ferrites [12-13], metals [14-15]) and various dielectrics such as ceramic, polymer, and other matrices [16]. Thermoreactive polymer composites containing silicon carbide [17-18], carbon fibers [19], carbon black [20], and carbon nanotubes [21-22] are also effective, which provide pronounced radio-absorbing and radio-shielding properties due to the various mechanisms of absorption combination: natural ferromagnetic resonance, motion resonance of domain boundaries, eddy current losses and repolarisation, multiple reflections, etc. Such materials effectively absorb electromagnetic radiation but have some disadvantages - synthesis complexity and high cost. Therefore, polymer composites for electromagnetic radiation radio absorption based on different thermoplastic matrices (polyvinyl butyral [23], polypropylene [24], polyvinylidene chloride [25]) with inorganic ferromagnetic fillers such as polymer ceramic-inorganic composites have been actively researched and designed. Receiving such materials is very perspective looking for their technological, operational and economic characteristics. In our previous works [26-27], the prospects of receiving polymer composites for absorbing electromagnetic radiation based on thermoplastic polyamide 6 and silicon carbide, which are relatively transparent in the millimetre frequency range and have a small absorption coefficient value, are shown. Our further research was aimed at the ceramic-inorganic polymer composites design using a complex fillers system: silicon carbide SiC, chromium oxide Cr₂O₃ and graphite (5-15% wt). Based on the scientific and technical papers which deal with materials for electromagnetic radiation radio absorption, the urgent task is to research a set of basic technological characteristics for creating effective products from them, as well as new approaches to modeling technological processes and designing products using composite polymer materials for electromagnetic radiation radio absorption.

The aim of the article is to study polymeric ceramic-inorganic composites' technological properties for absorbing electromagnetic radiation.

2. Experimental part

2.1. Raw materials

For the polymer ceramic-inorganic composites for electromagnetic radiation absorption, the following materials were used: - Durethane brand polyamide 6 (Bayer, Germany); - SiC silicon carbide powder (size - 50-65 microns); - Cr₂O₃ chromium oxide powder (size - 2-5 microns); - graphite powder (size - 20-25 μm), the produced from coke-chemical raw materials [28-29]. For the preparation of carbon graphite samples, standard graphite blanks were used, the photo of which is shown in Fig. 1. Table 1 shows the carbon graphite quality indicators.

Table 1. Carbon graphite quality indicators

Indicator	Symbol, measurement	Value
Total moisture	W^r_t , %	0.10
Analytical moisture	W^a , %	0.10
Ash	A^d , %	0.18
Carbon content	C^d , %	99.06
Hydrogen content	H^d , %	0.76



Fig. 1. Photo of graphite blanks

2.2. Experimental techniques

Polymer ceramic-inorganic composites for electromagnetic radiation absorption were received by extruding pre-prepared raw materials in a single-screw laboratory extruder at 170–200°C and a screw speed of 30–100 turns per minute

The melt flow index (MFI) study of polymer ceramic-inorganic composites was performed using the IIRT-M device at 270°C and a load of 2.16 kg.

The polymer ceramic-inorganic composites melting temperature was determined on a brass disk (diameter - 50 mm, thickness -19 mm, with a side hole for a thermometer with a diameter 9 mm). A brass disc equipped with a thermometer was gradually heated using a gas burner. Polymer samples measuring 10×10 mm was placed on it. Heating temperature intervals were recorded visually. The polymer ceramic-inorganic composites density was determined by the hydrostatic method, according to ISO 1183-1. The hydrostatic method is used to measure the polymer sample density with a volume of at least 1 cm³. The sample weighed in the air is attached with a thin wire to the rocker arm of the analytical balance, immersed in a beaker with distilled water or alcohol, placed on a special stand that does not touch the balanced cup, and weighed. Then the wire without the sample is weighed at the same immersion level.

The polymer density is calculated according to the formula:

$$\rho = m\rho_0 / \{m - (m_1 - m_2)\} \quad (1)$$

where m – sample's mass in air; g ; m_1 –the sample's mass with the wire in water; g ; m_2 –the wire's mass immersed in water or alcohol; g ; ρ_0 – water or alcohol density at 20°C.

3. Results and discussion

Primary studies were directed to study silicon carbide SiC, chromium oxide Cr₂O₃ and graphite introduction impact on polymer ceramic-inorganic composites technological properties. From previous works [26-27], it was found that increasing silicon carbide SiC in polyamide 6 led to an increase in both impact toughness and destructive stress during bending, when filler content is 5% wt. But overall protection against electromagnetic radiation efficiency of such received compositions was insufficient. So, in this work compositions with a higher silicon carbide content SiC - 10% wt were researched, and optimized the chromium oxide Cr₂O₃ and graphite content in the range of 5-15 % wt. were made. Polyamide 6 with 10% wt. silicon carbide SiC was used as based composite. Also, to found the synergistic effect of three different ceramic-inorganic graphite-ferromagnetic fillers integration, polyamide 6 - SiC - Cr₂O₃ - graphite systems were studied. In the table 2 filling degree of polymer ceramic-inorganic composites based on the polyamide 6 - SiC - Cr₂O₃ - graphite system impact on their technological characteristics is shown.

It can be seen (Table 2) that the introduction in polyamide 6 - SiC 10% wt. chromium oxide Cr₂O₃ and graphite (in different proportions) increase their melting temperature and density at the same time the MFI value is decrease. Due to graphite use there is an increase in the melting temperature from 220 to 223°C, density from 1195 to 1310 g/cm³, and a decrease in

MFI from 20 to 18.5 g/10 min. When using chromium oxide Cr₂O₃, the melting temperature increases from 220 to 221°C, the density from 1195 to 1305 g/cm³, and the MFI decreases from 20 to 19.5 g/10 min. With the simultaneous modification with chromium oxide Cr₂O₃ and graphite, the melting temperature increases to 224°C, the density reaches 1340 g/cm³, and the MFI decreases to 18.0 g/10 min. It can be concluded that binary composites modification with ceramic-inorganic graphite-ferromagnetic fillers is optimal for their technological characteristics:

- 1) polyamide 6 - SiC 10%wt - Cr₂O₃ 10% wt.
- 2) polyamide 6 - SiC 10% wt. - graphite 10% wt.

Table 2. Filling degree of polymer ceramic-inorganic composites based on the polyamide 6 - SiC - Cr₂O₃ - graphite system impact on their technological characteristics

Composition	Melting temperature, °C	Density, g/cm ³	MFI, g/10 min.
Polyamide 6 with 10% wt. silicon carbide SiC			
	220	1195	20.0
Polyamide 6 with 10% wt. silicon carbide SiC			
5 % wt. graphite	221	1225	19.5
10 % wt. graphite	222	1260	19.0
15 % wt. graphite	223	1310	18.5
Polyamide 6 with 10% wt. silicon carbide SiC			
5 % wt. Cr ₂ O ₃	221	1220	20.0
10 % wt. Cr ₂ O ₃	221	1255	19.5
15 % wt. Cr ₂ O ₃	221	1305	19.5
Polyamide 6 with 10% wt. silicon carbide SiC			
5 % wt. Cr ₂ O ₃ and 5 % wt. graphite	222	1260	19.0
10 % wt. Cr ₂ O ₃ and 10 % wt. graphite	224	1340	18.0

For three-component complex modification with ceramic-inorganic graphite-ferromagnetic fillers, the most optimal system is polyamide 6 - SiC 10% wt. - Cr₂O₃ 10% wt. - graphite 10% wt. For such systems, there are wide processing possibilities polymer ceramic-inorganic composites by injection moulding and extrusion methods to receive products, elements and parts- such as sheets, threads, meshes, tubes, screens- for electromagnetic radiation absorbing.

4. Conclusions

In the article the technological properties of polymeric ceramic-inorganic composites for absorbing electromagnetic radiation were studied. Modification effect for polymer based (polyamide 6) compositions with ceramic-inorganic graphite-ferromagnetic fillers on the melting temperature, density and melt flow index was determined.

The binary composites modification with ceramic-inorganic graphite-ferromagnetic fillers is optimal for their technological characteristics: 1) polyamide 6 - SiC 10%wt - Cr₂O₃ 10% wt.; 2) polyamide 6 - SiC 10% wt. - graphite 10% wt. For three-component complex modification with ceramic-inorganic graphite-ferromagnetic fillers, the most optimal system is polyamide 6 - SiC 10% wt. - Cr₂O₃ 10% wt. - graphite 10% wt. For the found optimal compositions of polymer ceramic-inorganic composites for electromagnetic radiation absorption, the processing possibility by injection moulding and extrusion to receive different configurations products and elements was proved.

Symbols

W^a	moisture contents, %;
W_t^r	total moisture, %;
A^d	ash content, %;
C^d	content of carbon, %;
H^d	content of hydrogen, %;
MFI	melt flow index, g/10 min

References

- [1] Yu WC, Zhang GQ, Liu YH, Xu L, Yan DX, Huang HD, Tang JH, Xu JZ, Li ZM. Selective electromagnetic interference shielding performance and superior mechanical strength of conductive polymer composites with oriented segregated conductive networks. *Chemical Engineering Journal*, 2019; 373: 556-564.
- [2] Rusly SNA, Matori KA, Ismail I, Abbas Z, Awang Z, Zulkimi MMM, Idris FM, Zaid MHM, Zulfikri ND. Microwave absorption properties of single- and double-layer coatings based on strontium hexaferrite and graphite nanocomposite. *Journal of Materials Science: Materials in Electronics*, 2018; 29: 14031-14045.
- [3] Bannikov LP, Miroshnichenko DV. Adjustment and Interpretation of Coefficients for Coal Tar Viscosity/Temperature Equations. *Petroleum and Coal*, 2023; 65 (1): 173-182.
- [4] Bilets D, Miroshnichenko D, Karnozhitskiy P. Gasification of coke-plant wastes. *Petroleum and Coal*, 2020; 63(3): 1121-1130.
- [5] Bilets D, Miroshnichenko D, Ryshchenko I, Rudniev V. Determination of material balance gasification of heavy coal tars with lignite and walnut shell. *Petroleum and Coal*, 2020; 63 (1): 85-90.
- [6] Wanasinghe D, Aslani F. A review on recent advancement of electromagnetic interference shielding novel metallic materials and processes. *Composites Part B: Engineering*, 2019; 176: 107207.
- [7] Rusly SNA, Ismail I, Matori KA, Abbas Z, Shaari AH, Awang Z, Ibrahim IR, Idris FM, Mohd Zaid MH, Mahmood MKA, Hasan IH. Influence of different BFO filler content on microwave absorption performances in BiFeO₃/epoxy resin composite. *Ceramics International*, 2020; 4: 737-746.
- [8] Stancu V, Galatanu A, Enculescu M, Onea M, Popescu B, Palade P, Aradoaie M, Ciobanu R, Pintilie L. Influences of Dispersions' Shapes and Processing in Magnetic Field on Thermal Conductibility of PDMS-Fe₃O₄ Composites. *Materials*, 2021; 14(13): 3696.
- [9] Zhao B, Wang S, Zhao C, Li R, Hamidinejad SM, Kazemi Y, Park CB. Synergism between carbon materials and Ni chains in flexible poly(vinylidene fluoride) composite films with high heat dissipation to improve electromagnetic shielding properties. *Carbon*, 2018;127: 469-478.
- [10] Menga F, Wanga H, Huang F, Guoa Y, Wanga Z, Huib D, Zhou Z. Graphene-based microwave absorbing composites: A review and prospective. *Composites Part B*, 2018; 137: 260-277.
- [11] Bibi M, Abbas SM, Ahmad N, Muhammad B, Iqbal Z, Rana UA, Khane SU. Microwaves absorbing characteristics of metal ferrite/multiwall carbon nanotubes nanocomposites in X-band. *Composites Part B: Engineering*, 2017; 114: 139-48.
- [12] Wang F, Wang X, Zhu JF, Yang HB, Kong XG, Liu X. Lightweight NiFe₂O₄ with controllable 3D network structure and enhanced microwave absorbing properties. *Scientific Reports*, 2016; 6: 37892.
- [13] Lv R, Kang F, Gu J, Gui X, Wei J, Wang K, Wu D. Carbon nanotubes filled with ferromagnetic alloy nanowires: lightweight and wide-band microwave absorber. *Applied Physics Letters*, 2008; 93: 223105.
- [14] Liu QH, Xu XH, Xia WH, Che RC, Chen C, Cao Q, He J. Dependency of magnetic microwave absorption on surface architecture of Co₂₀Ni₈₀ hierarchical structures studied by electron holography. *Nanoscale* 2015; 7: 1736-43.15.
- [15] Lv HL, Jia GB, Wang M, Shang CM, Zhang HQ, Du YW. Hexagonal-cone like of Fe₅₀Co₅₀ with broad frequency microwave absorption: effect of ultrasonic irradiation time. *Journal of Alloys and Compounds*, 2014; 615: 1037-42.
- [16] Tong XC. *Advanced materials and design for electromagnetic interference shielding*: CRC Press, Boca Raton: Engineering & Technology 2008.
- [17] Lisachuk G, Kryvobok R, Voloshchuk V, Lapuzina O, Zakharov A. Study of Technological Features of Celsian Ceramics Creation. *Proceedings of the 2021 IEEE 11th International Conference "Nanomaterials: Applications and Properties" (NAP 2021)*, 2021: P. 1-4.

- [18] Lisachuk G, Kryvobok R, Lapuzina O, Maystat M, Kryvobok N, Voloshchuk V, Gusarova I. To the problem of the creation of high-temperature radio-absorbing composite ceramic materials. Proceedings of the 2018 IEEE 8th International Conference on Nanomaterials: Applications & Properties (NAP-2018), 2018; 1: 01SPN41.
- [19] Shah A, Wang YH, Huang H, Zhang L, Wang DX, Zhou L, Duan YP, Dong XL, Zhang ZD. Microwave absorption and flexural properties of Fe nanoparticle/carbon fiber/ epoxy resin composite plates. Composite Structures, 2015; 131: 1132–41.
- [20] Al-Ghamdi AA, Al-Hartomy OA, A-Solamy FR, Dishovsky N, Malinova P, Atanasova P, Atanasovde N. Conductive carbon black/magnetite hybrid fillers in microwave absorbing composites based on natural rubber. Composites Part B: Engineering, 2016; 96: 231–41.
- [21] Hu JT, Zhao TK, Peng XR, Yang WB, Ji XL, Li TH. Growth of coiled amorphous carbon nanotube array forest and its electromagnetic wave absorbing properties. Composites Part B: Engineering, 2018; 134: 91–7.
- [22] Makarova TL, Geydt P, Zakharchuk I, Lahderanta E, Komlev AA, Zyrianova AA, Kanygin MA, Sedelnikova OV, Suslyayev VI, Bulusheva LG, Okotrub AV. Correlation between manufacturing processes and anisotropic magnetic and electromagnetic properties of carbon nanotube/polystyrene composites. Composites Part B: Engineering, 2016; 91: 505–12.
- [23] Zhu G, Cui X, Zhang Y, Chen S, Dong M, Liu H, Shao Q, Ding T, Wu S, Guo Z. Poly (vinyl butyral)/graphene oxide/poly (methylhydrosiloxane) nanocomposite coating for improved aluminum alloy anticorrosion. Polymer, 2019; 172: 415–422.
- [24] Lai H, Li W., Xu L., Wang X., Jiao H., Fan Z., Lei Z., Yuan Y. Scalable fabrication of highly crosslinked conductive nanofibrous films and their applications in energy storage and electromagnetic interference shielding. Chemical Engineering Journal, 2020; 400:125322.
- [25] Liang C, Hamidinejad M, Ma L, Wang Z, Park CB. Lightweight and flexible graphene/SiC-nanowires/ poly (vinylidene fluoride) composites for electromagnetic interference shielding and thermal management. Carbon, 2020; 156: P. 58–66.
- [26] Lebedev V, Kryvobok R, Cherkashina A, Bliznyuk A, Lisachuk G, Tykhomyrova T. Design and Research Polymer Composites For Absorption Of Electromagnetic Radiation. Proceedings of the 2022 IEEE 3rd KhPI Week on Advanced Technology (KhPIWeek), 2022: 1–4.
- [27] Lebediev VV, Kryvobok RV, Cherkashyna HM, Blyzniuk OV, Lisachuk HV, Voloshchuk VV. Rozrobka ta oderzhannia polimernykh kompozytiv dlia pohlynannia elektromahnitnoho vy-prominiuvannia. Vcheni zapysky Tavriiskoho natsionalnoho universytetu imeni V. I. Vernadskoho". Ceriia "Tekhnichni nauky", 2022; 5 (33(72)): 261–265. (In Ukrainian)
- [28] Bannikov LP, Miroshnichenko DV, Nesterenko SV, Balaeva YS, Fatenko SV. Influence of small coal particles in coking batch on coal-tar quality. Coke and Chemistry, 2019; 62 (3): 95–102.
- [29] Danilov AB, Verdibozhenko GS, Drozdnyk ID, Miroshnichenko DV, Kaftan YS, Golovko MB. Predicting the yield of coke-plant products on the basis of petrographic analysis. Coke and Chemistry, 2012; 55 (11): 419–422.

To whom correspondence should be addressed: professor Denis Miroshnichenko, National Technical University «Kharkiv Polytechnic Institute», 61002, Kirpychova str.2, Kharkiv, Ukraine, E-mail: dvmir79@gmail.com
Orcid: