ISSN 1027-5495. Functional Materials, 28, No.2 (2021), p. 279-286.
doi:https://doi.org/10.15407/fm28.02.279

High-strength aluminosilicate glass composite materials with special electrophysical properties

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Received July 29, 2020

For the development of compositions of high-strength protective materials, lithium-aluminum silicate glass-ceramic materials based on lithium disilicate crystals and spodumene crystals were chosen as the basis. The gradient type composite material included: a first layer with low conductivity (glass-ceramic material); the second layer with medium conductivity (glass-ceramic material and filler, silicon carbide grade 54C in an amount of 30 parts by weight per 100 parts by weight of glass); third layer with medium conductivity (thin layer of graphite,which is applied by aerosol method). It has been determined that for the developed composite materials, the formation of a gradient structure makes it possible to increase such electrical properties as tgδ from 0.005 to 0.03÷0.04; ε from 4.75 to 6.0÷7.0 and the strength properties KCU to 6.2 kJ/m^2 and K_{1C} up to 4.2 MPa·m$^{1/2}$.

Keywords: gradient type radio-absorbing glass-composite materials, dielectric constant, dielectric loss tangent, mechanical properties, armor elements.

Високоміцні алюмосилікатні склокомпозиційні матеріали зі спеціальними електрофізичними властивостями. *О.В.Саввова, Н.К.Блінова, О.І.Фесенко, Г.К.Воронов, О.В.Бабіч, С.О.Рябінін*

Досліджено склади композиційних високоміцних захисних матеріалів – літійалюмосилікатних склокристалічних матеріалів на основі кристалів дисилікату літію та кристалів сподумену. Композиційний матеріал градієнтного типу включав: перший шар з низькою провідністю (склокристалічний матеріал); другий шар з середньою провідністю (склокристалічний матеріал та наповнювач карбід кремнію у кількості 30 мас.ч. на 100 мас.ч. скла); третій шар з високою провідністю (тонкий шар графіту, який нанесено аерозольним методом). Визначено, що для розроблених композиційних матеріалів формування градієнтної структури дозволяє підвищити показники tgδ до 0,03, ε до 6,0÷7,0 та міцносні властивості KCU до 6,2 кДж/м^2 і K_{1C} до 4,2 МПа·м$^{1/2}$.

Исследованы композиционные высокопрочные защитные материалы – литийалюмосиликатные стеклокристаллические материалы на основе кристаллов дисиликата лития и кристаллов сподумена. Композиционный материал градиентного типа включал: первый слой с низкой проводимостью (стеклокристаллический материал); второй слой с средней проводимостью (стеклокристаллический материал и наполнитель і карбид кремния в количестве 30 мас.ч. на 100 масс.ч. стекла); третий слой со высокой проводимостью (тонкий слой графита, который нанесен аэрозольным методом). Определено, что для разработанных композиционных материалов формирование градиентной структуры позволяет повысить показатели tgδ до 0,03, ε до 6,0÷7,0 и прочностные свойства KCU до 6,2 кДж/м^2 и $К_{1С}$ до 4,2 МПа·м$^{1/2}$.

1. Introduction

The creation of modern high-strength functional materials with high physical and technical properties is a key stage in solving many engineering problems aimed at the development of complex equipment, in particular, computer technology, radio engineering, automation, radar, satellite communications, navigation, etc. The importance of the development of radio-absorbing materials is associated with the need to ensure the protection of information that can be lost through the channels of spurious electromagnetic radiation and to reduce the radar signature of various objects of weapons and military equipment. To date, a number of "Stealth technologies" have been developed and implemented — a complex of effective technical solutions aimed at reducing the level of signals coming from a military facility to receiving systems capable of detecting and destroying it [1]. It is the need to create technological high-strength radio-absorbing materials as armor elements that determines the priority of this direction for solving the main tasks of the country’s defense capabilities.

Most of the known radio-absorbing materials (RAM) are composites: gradient or interference. They consist of organic or inorganic (oxides and nitrides) substances, into which active absorbing components are introduced: powders of graphite, metals and carbides, ferrites or mixtures thereof, in particular, barium hexaferrites [2]. The matrix is made of a dielectric with the required electrical and mechanical parameters. From a physical point of view, RAMs are characterized mainly by thickness, coefficients of transmission (T) and absorption (R) of electromagnetic radiation and an operating wavelength range.

Today, radio-absorbing polyfunctional ceramic materials based on graphite with high thermomechanical and electrical properties for use in space have gained particular popularity [3]. One of the elements of the modern ceramic-based RAM technology is the implementation of the principles of the predicted microstructure. Thus, materials based on oxide systems $MgO–TiO_2–La_2O_3$, $SrO–TiO_2–MgO–ZnO$, $BaO–TiO_2–MnO_2$ with the presence of two or more crystalline phases have been developed. Crystalline phases with low dielectric losses form the basis of the matrix material ($MgTiO_3$), and compounds with high dielectric losses are $SrTiO_3$ crystallites. The oxides of the rare earth elements cerium and lanthanum were used as modifiers. When creating elements that absorb radio waves of high-frequency and ultra-high-frequency (microwave) ranges, high values of ε (dielectric constant) and tanδ (dielectric loss tangent) can be achieved; when strontium is replaced by bismuth in a RAM based on $SrTiO_3$ in a solid solution, and titanium is replaced by chromium, manganese or iron, a greater value of both ε and tanδ is achieved both at room temperature and at elevated temperatures. For radio-absorbing products, various compositions based on silicates, alumina silicates, aluminaborosilicates containing alkaline and alkaline earth oxides, which are characterized by lower synthesis temperatures and have better processability, can be used. The determining factor for their effective use is their ability to absorb electromagnetic radiation and high thermomechanical properties.

Recently, interest has increased in the use of glass-crystalline materials for the creation of radio-transparent and radio-absorbing products. The well-known radio-absorbing foam glass ceramics RPK-1 (LLC KERAPEN) (Table 1) based on natural raw materials — perlite, calcium borates and coke, is characterized by a low level of reflection of electromagnetic radiation from a flat surface [6]. For a long time, phased array antennas (PAR) based on ceramic material of the MTS-25 brand and glass-ceramic materials of the ST-38-1 and ST-32-1 brands of LLC "CDB RM" (Table 1), operat-

Table 1. Properties of well-known ceramic and glass-ceramic materials

Properties	Micro-wave ceramics	Microwave glass-ceramics		Radio-absorbing glass-ceramics	Radar-transparent glass-ceramics
	MTS-25	ST-38-1	ST-32-1	RPK1	OTM-357-U
ρ, kg/m^3	4700	2900	3100	430÷550	2410÷2550
CTE, $\alpha \cdot 10^7$, °C^{-1}	–	38	32	50	17
σ_{bend}, MPa	142	100	100	–	137
$\sigma_{compres}$, MPa	–	–	–	2.0÷4.50	–
λ, W/m·K	–	1.63	1.67	0.4÷0.06	2.3
ε, $f = 10^{10}$ Hz	25	7.25÷7.40	9.7÷10.0	–	6.5÷7.5
tgδ, $f = 10^{10}$ Hz	0.0005	0.0002	0.0003	–	0.015

ing in the microwave range, are successfully used in the serial production of radar stations [7]. However, in the known porous radio-absorbing glass materials, the mechanical properties deteriorate due to the significant content of the glass phase and the porosity of the material, which prevents their use under significant dynamic loads.

An effective solution in the development of radio-absorbing glass-ceramic materials is the use of the $SrO–Al_2O_3–SiO_2–B_2O_3$ system as a glass base [8] of magnitude, respectively, and significantly increase their crack resistance The introduction of carbon nanotubes and graphene into high-strength strontium-anortite glass-ceramics makes it possible to significantly reduce the electrical resistance of the clomatrix by 10 and 3 orders of magnitude. However, along with low values of dielectric characteristics and relatively high thermomechanical properties of strontium anorthite (temperature coefficient of linear expansion (CTE) $\alpha \cdot 10^7 = 26 \div 48$ deg^{-1}, elastic modulus $E = 100$ GPa, bending strength $\sigma_{bend} = 100 \div 120$ MPa), it has a sufficiently high melting temperature of 1710°C, which leads to an increase in the sintering temperature of glass ceramics to 1200°C, and the introduction of carbon nanotubes and graphene leads to a complication and increase in the cost of the technological process.

Glass-ceramics based on the systems $MgO–Al_2O_3–SiO_2$ (M–A–S) (9606, Corning, USA) and $Li_2O–Al_2O_3–SiO_2$ (L–A–S) (9608, Corning; OTM-357-B, RF) (Table 1) is characterized by a low level of radio waves reflection, a low coefficient of thermal expansion, temperature stability in the range of operating temperatures, low dielectric losses and high mechanical properties; it has been successfully used as protective radio-transparent elements of vehicles for a long time [9]. However, the well-known protective glass-ceramic materials are also characterized by rather high temperatures of melting (1450–1650°C) and crystallization (1200–1250°C), which significantly increases their cost and narrows the range of their application as replaceable armor elements [10, 11].

Glass-ceramic armor for viewing windows of military vehicles and helicopter windscreens based on lithium disilicate with high mechanical strength has high efficiency [12]. However, these materials are obtained under conditions of low-temperature heat treatment requiring long periods of the stage of crystal nucleation, which affects their processability and cost. High-strength lightweight glass-ceramics based on the L–A–S system, which contains 30–65 wt. % lithium disilicate and 20–60 wt. % β-spodumene [13], cannot be used under conditions of significant dynamic loads due to its low hardness.

Therefore, the creation of high-strength radio-absorbing glass-ceramic materials based on L–A–S systems for local protection with a simultaneous combination of high thermomechanical and electrical characteristics determines the relevance of this work and the need for further research.

2. Experimental

2.1. Purpose and methods of research

The main factor in the implementation of high protective properties against high-speed mechanical impact and electromagnetic radiation of armor elements based on glass-ceramic materials is to ensure the formation of high-strength ceramizing structure of a gradient-type composite material based on a dielectric matrix and a radio-absorbing component under conditions of short-term low-temperature heat treatment.

For radio-absorbing gradient-type materials, there is a gradual change in wave resistance and conductivity from values characteristic of free space in the first layer to low resistance and high conductivity of the final layer of the absorber; as a result, the maximum absorption coefficient is achieved. The minimum reflection coefficient is achieved provided that the value of the effective dielectric constant in the surface layer is minimally different from the free space and increases deep into the sample.

To solve this problem, it is necessary to develop high-strength radio-absorbing glass-composite aluminosilicate materials of the gradient-type and to study their operational properties, which was the purpose of this work.

The dielectric constant and dielectric loss tangent were calculated using the mathematical model developed by the authors [13]; the reflection coefficient and the coefficient of wave transmission from the waveguide diaphragm at $f = 10^{10}$ Hz and a temperature of 20°C were used. The mathematical model of this structure is based on the idea of representing the field in the cavity of the waveguide slot in the form of a field of natural waves in a waveguide equivalent to a slot. In the diaphragm with thickness t, a transverse slot with a length L and a width d is cut. The slot is partially filled with a dielectric — a developed material in the form of plates with $L = 5$ mm; d = 1.5 mm; $t = 2$ mm. The relative dielectric constant (ε) and dielectric loss tangent ($\tan\delta$) at the frequencies of ultrashort waves were determined using a computer comparative analysis of the dependence of the experimental and calculated frequency on the standing wave ratio (SWR) of the prototype. The relative permittivity (ε_r) was determined from the correspondence of the experimental resonance frequency to the calculated one. The dielectric loss tangent was determined from the correspondence of the values of the experimental SWR to the calculated ones.

The Vickers hardness and fracture toughness were determined by indenting the Vickers pyramid with a load of 5000 g for 5 measurements obtained on a TMV-1000 hardness tester. Impact toughness (KCU) was determined according to GOST 11067-2013 (EN1288-1:2000).

Young's modulus (E, GPa) of materials was determined on a "Sound-107" device by the method of determining the resonant vibration frequency with a relative error of ±0.2 % according to the formulas:

$$E = f_n^2 \cdot L^2 \cdot \rho;$$

$$f_n = n\frac{c_{sound}}{2L},$$

where n is the the number of harmonics, $n = 2$; L is the length of the sample; ρ is the density of material; c_s is the sound speed.

Petrographic studies of the materials were carried out using an optical polarizing microscope NU-2E.

2.2. Development of composite high-strength protective materials

For the development of formulations of composite high-strength protective materials, as the materials previously synthesized and investigated by the authors were chosen as the basis: lithium aluminosilicate glass-ceramics based on lithium disilicate crystals (SL series) and based on spodumene crystals (SP series) [14]. The glass-ceramic materials were obtained by two-stage low-temperature heat treatment and molded using ceramic technology by slip casting. The developed glass-ceramic materials are characterized by high values of Vickers hardness H (8.82÷8.90 MPa) and fracture toughness K_{1C} (3.15÷3.40 MPa·m$^{1/2}$), that is an important factor in the absorption of bullet impact energy without cracking and destruction.

A high-strength semiconductor material α–SiC was used as a filler, which was introduced into the original material in an amount of 10, 20 and 30 wt. %. It is known that α–SiC is used to create structures that absorb electromagnetic radiation and is characterized by a certain distribution of conductivity (σ), electric (ε) and magnetic (μ) permeability and a low wave reflection coefficient. Earlier, the authors of the work proved that the introduction of α–SiC into the composition of the developed materials in an amount of 10 wt. % per 100 wt. % leads to blocking of cracks and an increase in hardness due to structural rearrangement of the material [14].

Composite three-layer gradient-type materials were obtained on the basis of glass-ceramic materials SL-9 and SP-10 (first layer I) (Fig. 1); glass-ceramic materials SL-9 and SP-10 and a silicon carbide filler (grade 54C) in the amount of 10, 20 and 30 wt. % (second layer II) (Fig. 1). To achieve high conductivity, a thin graphite layer was formed by the aerosol method on the surface of glass-ceramic materials (third layer III)

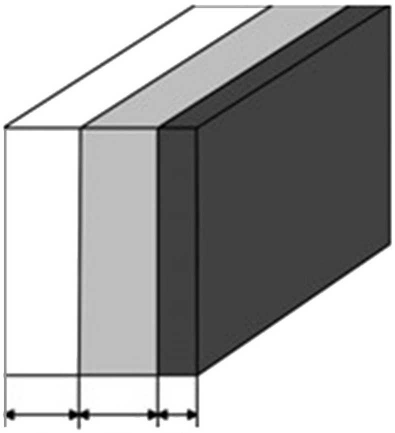

Fig. 1. The layer-by-layer structure of the composite material.

(Fig. 1); in general, this should significantly increase the absorption coefficient of the materials. Composite three-layer materials containing silicon carbide in the amount of 10, 20 and 30 wt. % were designated SL-9-KK-10, SP-10-UK-10, SL-9-KK-20, SP-10-UK-20, SL-9-KK-30, and SP-10-UK-30.

Samples in the form of a plate had the following dimensions: width — 2 cm, height — 1 cm and thickness — 2.1 mm; the thickness of the first layer is 1 mm, the second layer is 1 mm, the third layer is 0.1 mm. The layer-by-layer structure of the composite material is shown in Fig. 1.

Heat treatment of the composite materials was carried out under the same conditions as for the initial materials SP-10 and SL-9 at temperatures of 600 and 530°C, respectively, for 2 h (I stage), and at 900 and 850°C for 1.5 and 2.0 h, respectively, (II stage). This allows the formation of a high-strength fine-crystalline structure of the material and the formation of a protective layer of SiO_2 on the surface of SiC crystals, providing crack healing, increasing the fracture toughness of the composite and additional protection against high-temperature oxidation.

2.3. Study of the effect of structure on the performance properties of composite materials

The original glass-ceramic materials SL-9 and SP-10 are characterized by low values of dielectric losses (0.006 and 0.005, respectively) and dielectric constant (4.4 and 4.75) in the microwave range at a low temperature ($f = 10^{10}$ Hz; $t = 10°C$); the peculiarities of their structure are the presence of calcium, magnesium and aluminum cations in the glass phase, which block interatomic voids, and the presence of a significant amount of the β-spodumene crystalline phase with low dielectric losses ($\varepsilon = 6\div7$ at $t = 20°C$ and $f = 10^6$) [15]. This is due to the fact that the source of losses for glass-ceramics is the residual glass phase. The less of this phase in the glass-ceramics and the more it is free from alkali cations, the lower the dielectric losses. The low value of ε and tanδ in the prototypes at microwave frequencies is explained by a decrease in relaxation losses due to a decrease in the intermediate surface polarization; they are determined by ion and electronic polarization. The indicated losses for the glass-ceramic materials under study are low due to a low content of alkali metal ions in the glass phase [16]. The Li^+ cation, which has the greatest effect on losses and electrical conductivity in the structure of glass-ceramic materials SL-9 and SP-10, is included in the crystal lattice of lithium disilicate or β-spodumene and practically does not affect the increase in tanδ. The radio transparency of the developed glass-ceramic materials ensures the absence of interaction in the infrared, ultraviolet and visible ranges for the operation of on-board wireless communication systems.

For composite materials SL-9-KK-10 and SP-10-UK-10, the formation of a gradient structure (the second layer contains 10 wt.% α–SiC; the third layer is graphite)

Table 2. Properties of developed glass-ceramic materials

Properties	Developed glass-ceramic materials							
	SL-9	SL-10	SL-9. KK-10	SP-10. KK-10	SL-9. KK-20	SP-10. KK-20	SP-9. KK-30	SP-10. KK-30
KCU, kJ/m^2	4.9	5.0	5.7	5.7	6.0	6.1	6.2	6.2
K_{1C}, MPa·m$^{1/2}$	3.15	3.40	3.48	3.58	3.76	3.82	4.25	4.2
tanδ, $f = 10^{10}$ Hz	0.005	0.006	0.015	0.015	0.03	0.03	0.03	0.04
ε, $f = 10^{10}$ Hz	4.40	4.75	5.9	5.5	6.2	6.0	7.0	6.2

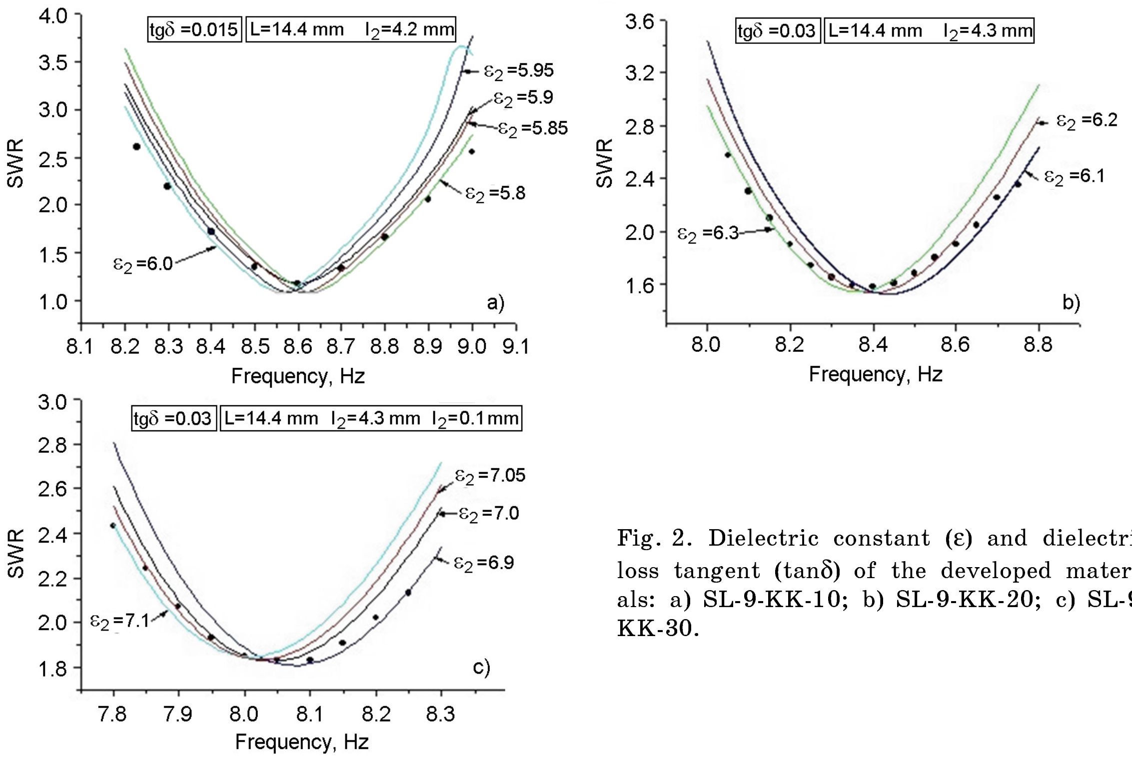

Fig. 2. Dielectric constant (ε) and dielectric loss tangent ($\tan\delta$) of the developed materials: a) SL-9-KK-10; b) SL-9-KK-20; c) SL-9-KK-30.

leads to a significant increase in the $\tan\delta$ indicator by a factor of 3 and in the ε indicator by 1.3 times (Table 2, Fig. 2a, 3a). An increase in the content of α–SiC to 20 and 30 wt. % in the second layer of composite materials SL-9-KK-20 SP-10-UK-20 and SL-9-KK-30 SP-10-UK-30 also leads to an increase in $\tan\delta$ to 0.03 and ε to $6.0\div7.0$ (Figs. 2b, 2c, 3b, 3c); this is an important condition for ensuring their ability to absorb waves in the microwave range at low temperatures. It should be noted that an increase in the ε indicator is also affected by an increase in the content of the crystalline phase, due to the introduction of α–SiC as a heterogeneous catalyst. In the future, an increase in the size of samples to real ones (up to 10×10 cm) will significantly increase both the $\tan\delta$ and ε indicators and the radio absorption capacity of materials.

For all the developed composite materials, the values of mechanical properties increase with increasing the content of α–SiC in the second layer from 10 to 30 wt. % (Table 2). With the maximum α–SiC content of 30 wt. %, there is an increase in impact strength and fracture toughness up to 6.2 kJ/m^2 and 4.2 MPa·m$^{1/2}$, respectively. This allows them to be used as armor elements to protect unarmored vehicles and makes them worthy competitors to ceramic materials (such as corundum, spinel), which are expensive and have a complex production technology. This is due to the fact that for composite materials SL-9-KK-20, SP-10-UK-20 and SL-9-KK-30, SP-10-UK-30 with an α–SiC content of 20 and 30 wt. % and for the materials SL-9-KK-10, SP-10-UK-10 with an α–SiC content of 10 wt. %, there is a tendency to an increase in the volume of the main crystalline phase, as for the initial glass-ceramic materials.

According to the results of petrographic analysis, an increase in the content of the crystalline phase up to 90 vol. % is observed in the composites. However, in this case, the structure of composite materials with a content of 10 wt. % silicon carbide, as well as with contents of 20 and 30 wt. % is finely dispersed. A further increase in the content of α–SiC up to 40 wt. % in the composition of the materials leads to weakening of their structure and, as a result, to deterioration of their mechanical properties (KCU = $1.0\div1.5$ kJ/m^2, K_{1C} = $0.5\div1.5$ MPa·m$^{1/2}$).

The developed composite materials, due to the specified combination of the characteristics of their layers, can be used as universal protective compositions. The first layer, which is a purely glass-ceramic material, plays the role of an energy-destroying and energy-absorbing layer. In this case, the modulus of elasticity for the developed materials is 308 GPa for SP-10 and 320 GPa for SL-9; this fact makes it possible, firstly,

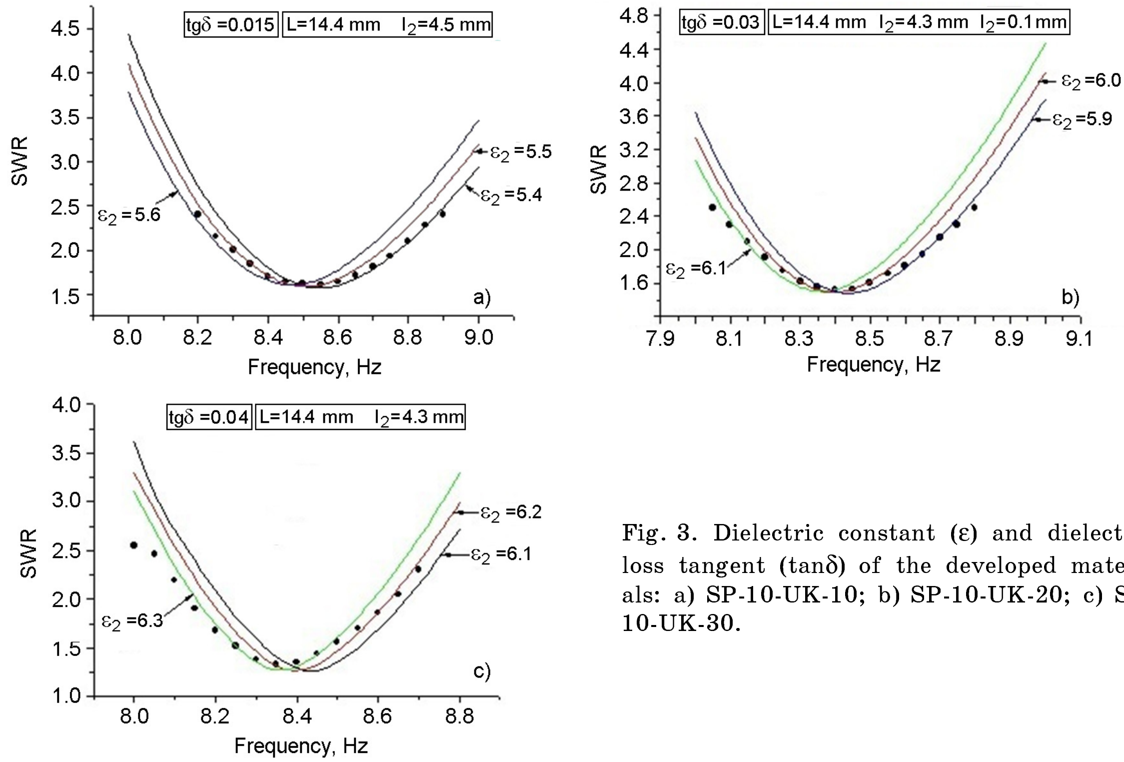

Fig. 3. Dielectric constant (ε) and dielectric loss tangent (tanδ) of the developed materials: a) SP-10-UK-10; b) SP-10-UK-20; c) SP-10-UK-30.

to provide energy absorption due to the presence of an amorphous phase, which removes mechanical microstresses caused by thermal factors and heals cracks caused by impacts; secondly, to destroy the damaging factor due to the significant speed of wave propagation in the obstacle.

The second glass-ceramic layer mainly acts as an energy-destructive layer due to the content of the α–SiC filler with high hardness and the presence of high-strength crystalline phases of spodumene or lithium disilicate in the amount of 90 vol.%. Due to the fragile destruction of the developed glass-ceramics into fragments up to 1 μm with the formation of radial cracks, the role of the crushing-deflecting layer increases. The presence of the third layer makes it possible to implement a mechanism for ensuring the minimum reflection coefficient of microwave radiation for the developed composite.

Creation of protection elements based on the developed gradient materials will provide a reliable shielding effect from microwave radiation and resistance to radio interference. Such materials, unlike traditional metal screens, have neither reflective effects nor negative impact on the shielded objects (equipment, people).

3. Conclusions

It has been established that the development of gradient-type composite materials based on aluminosilicate glass-ceramics, silicon carbide filler and sprayed graphite is promising for obtaining radio-absorbing protective materials for simultaneous protection against high-speed dynamic loading as well as for masking weapons and military equipment for observation using radar means.

Formation of a composite consisting of three layers (the first layer is a glass-ceramic material based on spodumene or lithium disilicate; the second layer is a glass-ceramic material based on spodumene or lithium disilicate with the addition of 20÷30 wt. % α–SiC; the third layer is graphite) made it possible to provide the main characteristics tanδ to 0.03 and ε to 6.0÷7.0, and to increase the ability to radio absorption of the material by increasing the dielectric constant from the first to the third layer in the depth of the sample.

For the developed composite materials, high impact strength (KCU = 6.2 kJ/m^2) and fracture toughness (K_{1C} = 4.2 MPa·m$^{1/2}$) were achieved due to the formation of a glass-ceramic structure containing high-strength crystalline phases of spodumene and lithium disilicate in an amount 90 vol. %, in particular, due to the content of 30 wt. % α–SiC in the second layer of the composition; these properties ensure the survivability of the armor elements during shelling and their operational en durance.

The obtained high-strength aluminosilicate glass-ceramic materials can be used in the manufacture of radio-absorbing armor elements for local protection of equipment and engineering facilities for military and civil purposes in the microwave range. The results of this work can be further used to develop materials for broadband radio absorbers in the form of radio-absorbing structures with a high level of protection against microwave radiation.

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