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DEVELOPMENT OF UNIVERSAL HIGH-STRENGTH MAGNESIUM ALUMINOSILICATE GLASS-CERAMIC MATERIALS FOR ARMOR PROTECTION

The necessity of creating innovative protective structures for armoring machinery and equipment is analyzed. A comparative analysis of high-strength materials used for local and individual body armor has been carried out. The relevance of the development of domestic universal armored materials that combine high strength properties and radio transparency based on lightweight technological magnesium aluminosilicate glass-ceramic materials with high performance properties at the level of world analogues has been determined. Were formulated the purpose and tasks of the work, which consist in: analysis of the current state of development of glass-ceramic materials for armor protection; development of magnesium-aluminosilicate glass-ceramic materials with high armor resistance, impact resistance and radio transparency; investigation of the operational properties of the developed glass-ceramic materials and their armor resistance; assessment of the competitiveness of the developed glass-ceramic materials. A set of requirements for a glass matrix for obtaining protective glass-ceramic materials has been formulated. Compositions and technological parameters for the production of glass-ceramic materials under the conditions of two-stage low-temperature heat treatment have been developed. The study of the electrical properties of experimental glass-ceramic materials made it possible to establish the possibility of effective functioning of on-board wireless communication systems when using the developed materials as elements of armor protection. It was found that the developed glass-ceramic materials are characterized by high operational properties (KCU = 5.8 kJ/m^2; HV = 10.4 GPa; K1C = 8.5 MPa·m$^{1/2}$) and armor resistance STANAG 4569 (level 2). The competitiveness of the developed glass-ceramic materials based on mullite is analyzed. It has been established that a significant increase in fracture toughness and a decrease in the density of the developed material when comparing properties with a similar armor element (corundum-based plate) will significantly reduce the weight of the armor elements by ≈1.4 times and its cost by five times, while maintaining its protective properties.

Key words: armor plate, glass-ceramic material, mullite, competitive ability.

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РОЗРОБКА УНІВЕРСАЛЬНИХ ВИСОКОМІЦНИХ МАГНІЙАЛЮМІНОСІЛІКАТНИХ СКЛОКЕРАМІЧНИМ МАТЕРІАЛІВ ДЛЯ БРОНЕЗАХИСТУ

Проаналізовано необхідність створення інноваційних захисних структур для бронювання техніки і обладнання. Проведено порівняльний аналіз високоміцних матеріалів, які застосовуються для локального та індивідуального бронезахисту. Визначено актуальність розробки вітчизняних універсальних бронематеріалів що поєднують високі міцносні властивості та характеристики радіопрозорості на основі полегшених технологічних магнійалюмосилікатних склокристалічних матеріалів з високими експлуатаційними властивостями на рівні світових аналогів. Сформульовано мету та завдання роботи, які полягають у аналізі сучасного стану розробок склокерамічних матеріалів для бронезахисту; розробці магнійалюмосилікатних склокристалічних матеріалів з високою бронестійкістю, ударостійкістю та радіопрозорістю; дослідженні експлуатаційних властивостей розроблених склокристалічних матеріалів та їх бронестійкості; оцінці конкурентної здатності розроблених склокристалічних матеріалів. Сформульовано комплексу вимог до скломатриці для одержання захисних склокристалічних матеріалів. Розроблено склади та технологічні параметри одержання склокерамічних матеріалів в умовах двостадійної низькотемператуної термічної обробки. Дослідження електричних властивостей дослідних склокристалічних матеріалів дозволило встановити можливість ефективного функціонування бортових безпровідних систем зв'язку при застосуванні розроблених матеріалів як елементів бронезахисту. Встановлено, що розроблені склокристалічні матеріали характеризуються високими експлуатаційними властивостями (KCU = 5,8кДж/м^2; HV = 10,4 ГПа; K_{1C} = 8,5 МПа·м$^{1/2}$) та бронестійкістю STANAG 4569 (2 рівень). Проаналізовано конкурентоздатність розроблених склокерамічних матеріалів на основі муліту та встановлено, що вагоме підвищення в'язкості руйнування та зниження щільності розробленого матеріалу при порівнянні з аналогічною пластиною на основі корунду дозволить суттєво знизити вагу бронеелементів у ≈ 1,4 рази та його вартість у 5 разів при збереженні його захисних властивостей. Визначено економічний ефект від впровадження вітчизняних склокерамічних матеріалів для бронезахисту.

Ключові слова: бронеелемент, склокерамічний матеріал, муліт, конкурентна здатність.

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РАЗРАБОТКА УНИВЕРСАЛЬНЫХ ВЫСОКОПРОЧНЫХ МАГНИЙАЛЮМИНОСИЛИКАТНЫХ СТЕКЛОКЕРАМИЧЕСКИХ МАТЕРИАЛОВ ДЛЯ БРОНЕЗАЩИТЫ

Проанализирована необходимость создания инновационных защитных структур для бронирования техники и оборудования. Проведен сравнительный анализ высокопрочных материалов, применяемых для локального и индивидуальной бронезащиты. Определена актуальность разработки отечественных универсальных бронематериалов которые объединяют высокие прочностные свойства и радиопрозрачность на основе облегченных технологических магнийалюмосиликатних стеклокерамических материалов с высокими эксплуатационными свойствами на уровне мировых аналогов. Сфоромулировано цель и задачи работы, которые заключаются в: анализе современного состояния разработок стеклокерамических материалов для бронезащиты; разработке магнийалюмосиликатных стеклокерамических материалов с высокой бронестойкостью, ударопрочностью и радиопрозрачностью; исследовании эксплуатационных свойств разработанных стеклокерамических материалов и их бронестойкости; оценка конкурентной способности разработанных стеклокерамических материалов. Сформулировано комплекса требований к стекломатрице для получения защитных стеклокерамических материалов. Разработаны составы и технологические параметры получения стеклокерамических материалов в условиях двухстадийной низкотемпературной термической обработки. Исследование электрических свойств опытных стеклокерамических материалов позволило установить возможность эффективного функционирования бортовых беспроводных систем связи при применении разработанных материалов как элементов бронезащиты. Установлено, что разработанные стеклокерамические материалы характеризуются высокими эксплуатационными свойствами (KCU = 5,8 кДж/м^2; HV = 10,4 ГПа; K_{1C} = 8,5 МПа·м$^{1/2}$) и бронестойкостью STANAG 4569 (2 уровень). Проанализирована конкурентоспособность разработанных стеклокерамических материалов на основе муллита и установлено, что значительное повышение вязкости разрушения и снижения плотности разработанного материала при сравнении свойств с аналогичным бронеэлементом (пластиной на основе корунда) позволит существенно снизить вес бронеэлементов в ≈ 1,4 раза и его стоимость в 5 раз при сохранении его защитных свойств. Определен экономический эффект от внедрения отечественных стеклокерамических материалов для бронезащиты.

Ключевые слова: бронеэлемент, стеклокерамический материал, муллит, конкурентная способность.

Introduction.

For Ukraine, the importance of the urgent problem of creating new protective materials has significantly increased recently in connection with the conduct of hostilities in the ATO zone. Currently, to protect special equipment and technology, there are no reliable shock-resistant armor elements of domestic production with a reduced cost, density and high manufacturability compared to existing foreign ceramic armor materials. The existing composite armor elements based on ceramic, polymer materials and metal alloys are an effective means of individual and local protection (transport, engineering structures, equipment) from high-speed dynamic load (debris, ball). However, the use of armor elements based on corundum (Al_2O_3) and functionally-graded ceramic materials based on composites «steel–corundum–steel» or Al_2O_3, which is hardened by ZrO_2, is limited by their high density and weight [1–4], and ceramics based on B_4C, B_4Ti, SiC – high cost and complexity of the production process [5]. An effective solution to this problem is the replacement of a destructive layer based on ceramics in the composition of composite armor elements with a destructive and energy-absorbing layer based on glass-ceramic materials, which are distinguished by a reduced cost and high operational properties. In addition, these glass-ceramic materials can have unique properties – they can be radio- and optically transparent, which allows them to be used as independent protective elements for special machinery and equipment. All this determines the relevance of creating a new type of universal domestic armored elements, characterized by a combination of high armor resistance and radio transparency. This will ensure reliable protection of military and civilian personnel and the operation of military equipment and technology, especially in combat conditions.

Literary review.

The widespread use of composite armor elements based on glass-ceramic materials (magnesium (MAS) and lithium aluminum silicate (LAS) systems) as replaceable elements with a high degree of optical and radio transparency is limited by high production temperatures, the cost of raw materials (LAS) and the complexity of the technological mode (MAS) [6–9]. Promising for the protection of optical devices of small arms is the use of domestic glass-ceramic materials based on lithium disilicate (LS_2), which are distinguished by the simultaneous combination of high mechanical strength to ensure resistance to the action of energy-destroying components and the ability to absorb and dissipate shock loads [10]. Also known is a high-strength glass-ceramic material for personal protection based on β-spodumene of the following composition, wt. % SiO_2 60,0; Al_2O_3 15.0 – 18.0; Li_2O 7.0 – 10.0; CaO 0.0 – 2.0; MgO 1.0 – 3.0; TiO_2 0.0 – 2.0; ZrO_2 0.0 – 1.0; ZnO 3.0 – 4.0; SnO_2 0.0 – 1.0; P_2O_5 3.0; B_2O_3 1.5 – 2.0; CeO_2 0.5; MnO_2 0.0 – 2.5 [11]. The developed material, due to the structural features, is characterized by low density indices ρ = 2.7 kg m^3 and temperature coefficient of linear expansion $\alpha \cdot 10^7 = 21.34$. However, insufficiently high indicators E = 95.6 GPa, K_{1C} = 3.4 MPa·$m^{1/2}$, HV = 8680 MPa do not allow the use of this material as high-strength for elements of soft-skinned armored vehicles. A significant obstacle to the use of lithium-containing glass-ceramic materials as armor elements is their increased cost due to the significant content of lithium oxide. Most foreign developments for this system refer to transparent or translucent sitalls, which have low mechanical properties [12].

The efficiency of using cordierite glass-ceramic materials as protective elements is determined by their extremely high mechanical and thermal properties. There is a composition of glass-ceramic composite for armor protection, which contains alumina and glass-ceramic material, which is obtained by the method of liquid-phase sintering [4]. This glass-ceramic was obtained on the basis of the $MgO–Al_2O_3–SiO_2$ system and had a composition (in wt. %): Al_2O_3 – 29.07; Na_2O – 0.17; Fe_2O_3 – 0.20; MgO – 15.83; SiO_2 – 43.25; CaO – 0.69; K_2O – 0.19, characterized by the presence of α-cordierite after firing. The use of glass-ceramics in the composition of the composite in an amount of 7 or 14 wt. % allows one to reduce the time and temperature of composite sintering from 1670 °C for 5 h to 1570 °C for 0.5 h and obtain a composite with a density of ρ = 2.7 kg/m^3, hardness HV = 10.8 GPa and modulus of elasticity E = 310–343 GPa. However, the fracture toughness of the composite (K_{1C} = 2.50–3.85 MPa·$m^{1/2}$) is not enough to ensure reliable survivability of armor during shelling and operational survivability.

The use of transparent glass-ceramic materials based on the $MgO–Al_2O_3–SiO_2$ system as transparent protective structures is widely known. Transparent glass-ceramic material containing, by wt. %: SiO_2 – 48.41; Al_2O_3 – 26.15; MgO – 9.74; TiO_2 – 8.00; ZrO_2 – 0.89; B_2O_3 – 1.52; Sb_2O_3 – 5.29 is obtained by a two-stage heat treatment mode: the first stage (I st.) at a temperature (T) of 727 °C, during (τ) 5 hours; the second stage (II st.) at T = 900 °C for τ = 3 h. Long exposure at the stages of nucleation of quartz (I st) and spinel (II st) allows to provide high values of HV = 9880 MPa. However, they have insufficiently high indicators of mechanical properties E = 105 GPa, K_{1C} = 3.0 MPa·$m^{1/2}$ [8].

Therefore, the urgent task of increasing the reliability of the protection of special equipment and equipment, which is operated under conditions of high temperatures and mechanical loads, is the development of domestic universal armor materials that combine high strength properties and radio penetration, based on lightweight technological magnesium aluminum silicate glass-ceramic materials with high operational properties at the level world analogues.

The aim of work

The aim of this work is to develop universal high-strength magnesium aluminosilicate glass-ceramic materials for armor protection.

To achieve it, the following tasks were set:

– analysis of the current state of development of glass-ceramic materials for armor protection;

– development of magnesium aluminosilicate glass-ceramic materials with high armor resistance, impact resistance and radio transparency;

– research of operational properties of the developed glass-ceramic materials and their bullet-resistant protection;

– assessment of the developed glass-ceramic products competitiveness.

The results of the experiment and their discussion.

To obtain protective glass-ceramic materials, the glass matrix must be in conformity with following set of requirements: the ability to finely dispersed bulk crystallization with the formation of high-strength crystalline phases (α-cordierite, spinel, mullite) in a two-stage thermal treatment; density below 3.0 g/cm^3; high heat and fire resistance.

To establish the area of existence of the initial glasses, the system was chosen K_2O–RO–RO_2–P_2O_5–R_2O_3–SiO_2. It was limited to the following concentration limits, wt. %: K_2O 2.0 – 7.0; MgO 9.0 – 14.0; CaO 2.0 – 5.5; ZnO 2.0 – 2.5; SrO 2.0 – 4.0; TiO_2 2.0 – 8.0; ZrO_2 2.0 – 2.5; CeO_2 0.0 – 0.5; Al_2O_3 20.0 – 30.0; B_2O_3 0.0 – 5.0; P_2O_5 0.0 – 3.0; SiO_2 45.0 – 53.0.

In laboratory conditions, 10 compositions of magnesium-aluminosilicate glasses with the KGC marking were made, which were proposed for obtaining a glass-ceramic material. A comparative assessment of the properties of the developed glass-ceramic materials (table 1) and well-known ceramic and glass-ceramic materials, which are widely used as armor elements, has been carried out [12]. All experimental glasses were melting under the same conditions at a melting temperature (T_{ml}) of 1550–1600 °C in corundum crucibles, followed by cooling on a metal sheet. The marking of glass-ceramic materials corresponds to the marking of the glasses on the basis of which they were obtained. Glass-ceramic materials based on glasses were obtained by ceramic technology with a two-stage heat treatment mode: heat treatment temperature (Tt) stage I – 780–850 °C, for 1 hour, stage II – 1050–1150 °C, for 1 hour. When designing the compositions of research glasses for obtaining armored elements, one of the main factors is to ensure the ability to form at the stage of nucleation (I st), by the mechanism of phase separation, nanostructures under conditions of low-temperature processing for a short period. Subsequent heat treatment of such samples (II st) should ensure the occurrence of crystallization with the presence of an interconnected structure, which determines the high operational properties of glass-ceramic materials and the possibility of their use under conditions of significant mechanical and thermal loads.

Table 1 – Properties of the developed glass-ceramic materials and known [12] ceramic and glass-ceramic materials for armor protection

Marking of materials		Properties								
		H, GPa	*HV*, GPa	K_{1C}, MPa·m$^{1/2}$	*E*, GPa	*KCU*, kJ/m^2	$\alpha \cdot 10^7$, °C^{-1}	ρ, g/cm^3	tgδ·10^4	ε
Corundum ceramics AD-995 Industrial Ceramic Materials		–	18.00	4.00–5.00	370	–	–	3.90	–	–
Boron carbide		–	28.00	3.70–4.30	475	–	–	2.40–2.52	–	–
Silicon carbide		–	20.00	2.90–3.50	330–370	–	–	3.00	–	–
Spinel glass-ceramics		–	9.88	3.00	105	–	–	2.43	–	–
Developed materials	CGC-1	9.20	9.40	3.50	120.6	5.0	48.9	2.63	70	7.5
	CGC-2	9.20	9.30	3.30	110.5	4.0	43.8	2.47	72	7.6
	CGC-3	9.50	9.55	3.50	140.7	5.0	31.8	2.50	5	5.5
	CGC-4	9.35	9.40	3.40	130.4	4.2	45.7	2.49	10	6.0
	CGC-5	9.40	9.45	3.40	138.9	4.8	37.2	2.48	8	6.0
	CGC-6	9.20	9.30	3.30	115.7	4.7	44.8	2.47	58	6.5
	CGC-7	9.20	9.40	3.30	120.9	4.6	36.8	2.47	22	6.8
	CGC-8	9.50	9.60	3.40	300.2	5.2	38.7	2.74	55	4.0
	CGC-9	9.60	9.80	3.50	340.8	5.3	44.8	2.76	55	4.0
	CGC-10	9.60	9.80	3.50	350.6	5.5	57.7	2.80	50	3.9
	CGC-10-ZD-5	10.00	10.40	8.52	360.2	5.8	56.5	2.82	46	4.0

Investigation of the electrical properties of experimental glass-ceramic materials of the CGC series made it possible to establish the following features. At frequencies of 10^{10} Hz for test materials, fixing the dielectric loss tangent index tgδ = 0.0003–0.0050 (Tab. 1) indicates a decrease in relaxation processes (dielectric losses with pronounced relaxation polarization sharply decrease when going from 10^7 to 10^{10} Hz). This is especially important for glass-ceramic dielectrics based on mullite or cordierite, the crystalline phase of which consists of substances that are characterized by a structure with loose packing of ions and have increased dielectric losses caused by relaxation polarization.

Low tgδ values at a current frequency of 10^{10} Hz associated with the satalization of the structure of experimental materials, which consists in the formation of a significant number of fine crystals and their mutual penetration, which is accompanied by the leveling of the gradient of properties at the phase boundaries, a decrease in the contact area and the boundaries of surface polarization. Thus, the smallest tgδ value is observed for the CGC-3 material, and the highest for the CGC-1, CGC-2 and CGC-4 materials, which is associated with a decrease in the content of the crystalline phase of α-cordierite in them. The tgδ ≈ 0.005 value for glass-ceramic materials CGC-8, CGC-9, CGC-10 will ensure the effective functioning of on-board wireless communication systems (lack of interaction in the infrared, ultraviolet and visible ranges). Ensuring the radio transparency of the experimental materials CGC-8, CGC-9, CGC-10 at the level of ε ≈ 4.0 will increase the thickness of the armor element to increase the ballistic resistance of the equipment. The stability under the action of an open flame for test materials is determined by their high heat resistance. The value of the thermal coefficient of linear expansion (TCLE) for glass-ceramic materials is determined by the content: for samples with CGC-1 to CGC-7 – α-cordierite and is the lowest for CGC-3 material with a content of the indicated crystalline phase of 75 vol. %; for glass- ceramic materials CGC-10, CGC-9 and CGC-8 – mullite content. The presence of aluminum-magnesium spinel, characterized by a high TCLE (80–97 · 10^7 °C^{-1}), leads to the appearance of stresses at the interfaces and increases the TCLE value of the CGC-1 material.

For experimental glass-ceramic materials CGC-1, CGC-2, CGC-3, CGC-4, CGC-5, CGC-6 and CGC-7, the strength properties are in direct proportion to the content of the crystalline phase of α-cordierite (HV = 8.2 MPa, K_{1C} = 2.3 MPa·$m^{1/2}$, E = 139 GPa). Thus, the highest values of Vickers hardness (HV) and fracture toughness (K_{1C}) are observed for the CGC-3 composition with a high content of α-cordierite, and the lowest values are for the CGC-2 and CGC-6 warehouses, respectively, with a low content of α-cordierite. For samples CGC-8, CGC-9 and CGC-10, which are characterized by a significant content of mullite (HV = 11.0 MPa, K_{1C} = 2.6 MPa·$m^{1/2}$, E = 220 GPa), a significant increase in HV, K_{1C} and impact strength (K_{CU}) (tab.1). For the density indicator, a similar trend is also observed, however, a slight increase in ρ is not a determining factor for ensuring the mobility of armored vehicles. It is extremely important to solve the problem of decreasing the tendency to fracture at "critical" loads. To create a material structure that would include elements that prevent crack propagation, change its trajectory, or blunt (block) the crack tip, thereby reducing the stress concentration, 5 wt. % yttrium-stabilized zirconia was introduced into the composition of composite materials. The developed glass-ceramic material CGC-10-ZD-5 is distinguished by high performance characteristics. The carried out ballistic tests showed that the developed glass-ceramic material CGC-10-ZD-5 withstood shelling according to the requirements of STANAG 4569 (level 2).

Comparative evaluation of the developed glass-ceramic material KSK-10-DC-5 and well-known representatives of ceramic armor materials made it possible to establish that the developed material is somewhat inferior to ceramic armor elements in terms of elasticity and hardness (tab. 2). However, a significant increase in fracture toughness and a decrease in the density of the developed material when compared with a corundum-based plate of a similar thickness will significantly reduce the weight of the armor element by ≈1.4 times and its cost by 5 times while maintaining its protective properties. An important aspect of the effective implementation of the developed element is its reduced cost due to the use of cheaper raw materials and simplified technology. The introduction of the developed high-strength glass-ceramic materials will increase the competitiveness of domestic armored materials and provide indicators of their properties at the level of world analogues.

Table 2 – Comparative characteristics of the density and cost of known ceramic and glass-ceramic armored materials and the developed glass-ceramic material

Material (basic system, manufacturer or developer, country of origin)	Density, g/cm³	Cost raw materials, USD/kg	The cost of the product, USD/kg
Boron carbide (CarSIK-B_4C, by Schunk Group, Germany)	2.50	26–30	300–1000
Silicon carbide (SiC, Hexoloy® by Saint-Gobain, USA)	3.20	10–20	300
Corundum (Alumina ballistic armour, Morgan Advanced Materials, England)	3.90	8–10	150
Spinel Transparent Armor (Aluminum-magnesium spinel, CeraNova Corporation, USA)	6.50	10–12	40
Aluminum oxynitride (ALON Transparent Armor, Surmet Corporation, USA)	7.00	20–25	60
Glass-ceramics (LAS glass ceramics, Elan Technology, USA)	2.35	2–5	30
Glass-ceramics CGC-10-ZD-5 (MAS glass ceramics, ХНУМГ ім. О.М. Бекетова, UA)	2.80	0.5–1.0	28

Conclusions.

The prospects for creating domestic armor elements based on high-strength glass-ceramic materials have been determined. The main provisions of the technology for obtaining protective glass-ceramic materials are formulated and the compositions of composite materials based on glass-ceramic magnesium-aluminosilicate materials and a filler – zirconium dioxide under conditions of two-stage low-temperature heat treatment are developed. It was found that the developed glass-ceramic materials are characterized by high operational properties (K_{CU} = 5.8 kJ/m^2; HV = 10.4 GPa; K^{1C} = 8.5 MPa·$m^{1/2}$) and armor resistance (level 2) and can be used as a basis when developing a composite element of body armor. The introduction of the developed materials as protective structures in the armoring of machinery and equipment will make it possible to solve the strategically important problem of providing the defense industry with reliable competitive domestic materials.

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