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TECHNOLOGY ORGANIC AND INORGANIC SUBSTANCES

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Designing new materials with unique properties requires scientifically substantiated approaches to problem-solving. Applying a physical-chemical analysis of oxide systems to devise the formulation for a material makes it possible to determine the conditions of phase formation and assess the manufacturability of compositions. Given the enormous number of experiments required to build the state diagrams of multi-component oxide systems, the physical-chemical modeling is the most appropriate method to study their structure. This paper substantiates the selection of the basic oxide system ZnO-SrO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> to design radio-transparent ceramics and reports the results of studying its subsolidus structure using modern data on splitting the system into elementary volumes. The main geometric-topological characteristics of the system's internal tetrahedra have been defined and analyzed; the minimum temperatures for melt occurrence have been calculated, as well as the eutectic compositions. To design radio-transparent ceramics with a predefined level of dielectric characteristics ( $\varepsilon < 10$ , tg  $\delta < 10^{-2}$ ), a region of the formulations has been selected within the tetrahedron  $SiO_2$ -ZnAl<sub>2</sub>O<sub>4</sub>-ZnSiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> concentrations, which ensure the synthesis of the target phases of willemite and strontium anorthite. By using the new data, heat-resistant polyphase ceramics have been obtained, whose dielectric characteristics ( $\epsilon$ =5.98-8.96; tg  $\delta$  =0.004-0.008) meet the requirements for radio transparent materials. The optimal ratio of phases (ZnSiO<sub>4</sub>:SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>=1:1) has been established, which makes it possible to reduce dielectric permeability ( $\varepsilon$ =5.98) and minimize dielectric losses (tg  $\delta$  =0.004). Scanning electron microscopy and X-ray analysis were used to determine the structural and phase features of the new ceramic materials

Keywords: subsolidus structure, geometric-topological characteristics, willemite, strontium anorthite, radio transparent ceramics

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#### 1. Introduction

The development of modern aviation has significantly increased the maneuverability and flight speed of aircraft (AC), as well as opened up opportunities for their all-weather use. That necessitated the need to protect external radio-electronic systems against external influences during AC operation. The main task in the manufacture of fairings is to choose such materials that would be suitable for use under extreme conditions of simultaneous exposure to thermal and aerodynamic loads, temperature fluctuations, and aggressive environments of high-speed gas flows.

Materials for the manufacture of fairings should not alter the amplitude of a passing electromagnetic wave and lead to a chaotic change in its phase [1]. The key requirement for UDC 666.651

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# SUBSOLIDUS STRUC-TURE OF THE ZnO-SrO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> SYSTEM AS A BASE FOR DESIGNING RADIO-TRANSPARENT CERAMICS

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such materials is radio transparency: the rate of reflection of radio waves should not exceed 1 %. In addition, the dielectric losses by radio-transparent materials (RTM) in the interval of working temperatures should be minimal (the tangent of the angle of dielectric losses is tg $\leq$ 0.01) while dielectric permeability is strictly standardized ( $\epsilon$ =1–10).

The specified requirements for the functional properties of RTM exclude the possibility of using many structural materials, in particular metals and fiberglass, for the manufacture of fairings. Composite polymer-based RTM, used to protect land- and sea-based antenna systems, are designed to operate in temperate temperatures (up to 500 °C). It also excludes them from the list of materials suitable for the manufacture of fairings for the modern supersonic AC and rocketry [2]. The increased requirements for materials for the manufacture of AC fairings determine the relevance of designing thermally resistant RTM with high strength, a service temperature of at least 1,200 °C, and the dielectric characteristics that are stable in the interval of working temperatures.

The quality of fairings and, as a result, reliable protection of radio-technical systems, requires not only the improvement of existing RTM technologies but also such new high-performance materials that should be developed on the basis of the fundamental knowledge about phase changes and equilibrium phase combinations, both in the synthesis and under operating conditions.

### 2. Literature review and problem statement

Constructing materials for the manufacture of fairings, which would possess a set of functional properties and could withstand high operating temperature, is given much attention. Among the existing high-temperature RTM, of interest are sitalls based on aluminosilicates (strontium anorthite, cordierite, celsian) [3, 4], as well as special types of ceramics such as quartz, nitride-silicon, high-alumina, etc. [5]. However, each specified material has its drawbacks. For example, the use of radio-transparent sitalls is limited by their heat resistance (a deformation temperature is 900–1,000 °C); therefore, the issue of stability of the dielectric characteristics of sitalls in the interval of the temperature of fairing operation is debatable. In addition, manufacturing fairings from sitalls [5, 6] has a series of flaws associated with high energy intensity as the temperature of glass melting reaches 1,650 °C. The obstacle to its widespread application is also the imperfection of article molding technology due to the instability of the rheological properties of slips, the low strength of cast billets, and the difficulty of extracting them from the molds.

Unlike the production of sitalls, ceramic technologies make it possible to obtain materials with a densely baked or regulated porous structure, as well as to ensure the reproducibility of their properties. In addition, radio-transparent ceramic materials demonstrate the stability of phase composition and dielectric characteristics at high temperatures. However, each of them has its advantages and disadvantages [7]. Thus, highly heat-resistant quartz ceramics is limited by the operational temperature of 1,000 °C [7,8]. High-alumina ceramics that demonstrate high strength and resistance to the effects of aggressive environments, have insufficient heat resistance (200 °C) and require high firing temperatures (1,600-1,700 °C) [9]. Silicon-nitride- and boron-based ceramics have a high melting point (3,000 °C), mechanical strength (flexural strength  $\sigma_f = 800 - 1,200$  MPa), heat resistance and, most importantly, high thermostability of dielectric characteristics (up to 2,000 °C). However, the production of articles from these materials is characterized by high energy intensity and complexity of thermal equipment: the powders of  $Si_3N_4$  and BN are baked by a hot-pressing method at temperatures of 1,600–1,700 °C and 1,800–1,900 °C, respectively [10].

At present, the specialists' efforts are aimed at obtaining effective radio-transparent ceramic materials with a reduced firing temperature. The authors of [11] devised a technology for manufacturing radio-transparent materials based on strontium anorthite with a  $SrAl_2Si_2O_8$  synthesis temperature of 1,250 °C with the article firing temperature of 1,350 °C. A given technology enables obtaining densely baked RTM (water absorption is W=0.08%) with an operating temperature of up to 1,500 °C and satisfactory dielectric characteristics ( $\varepsilon$ =4.93–5.26; tg $\delta$ =0.0097–0.0122 in the frequency interval of 26–37.5 GHz). Despite the undeniable advantages of the properties of strontium-anorthite ceramics, a given technology of fairing manufacturing is distinguished by complexity and multiple stages. Work [12] reports the results of designing radio-transparent ceramics based on the compositions of the SrO–BaO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> system. As a result of firing at 1,550 °C, the authors obtained the materials based on a solid solution of strontium and barium anorthite (SrO<sub>0.5</sub>BaO<sub>0.5</sub>Al<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>) whose dielectric permeability indicator is  $\varepsilon$ =9.2 and the reflection factor of radio waves is *S*=0.253.

The authors of study [13] designed polyphase radio transparent ceramics using ceramic technology in combination with the principle of the reactionary structure formation. The target phases of the ceramics are spodumene and cordierite, the combination of which ensures high heat resistance (at least, 1,050 °C) and satisfactory dielectric characteristics of the material ( $\varepsilon$ =4.93–5.26; tg $\delta$ =0.0097–0.0122 at a frequency of 10<sup>10</sup> Hz). While emphasizing the high technical characteristics of given ceramics, we should pay attention to the features of the technology that includes a three-stage high-temperature heat treatment: glass melting (1,350 °C), obtaining "cordierite fireclay" (1,200 °C), and firing the articles (1,300–1,350 °C). Although the resulting cordierite-spodumene ceramics are characterized by a lower firing temperature, the proposed technology is complex and energy-intensive.

In work [14], the polyphase ceramics with a composition of  $(1-x)(0.75\text{ZnAl}_2\text{O}_4-0.25\text{TiO}_2)-x\text{SrAl}_2\text{Si}_2\text{O}_8$  were obtained and studied. It is shown that varying the ratio of phases of gahnite, rutile, and strontium anorthite makes it possible to adjust the dielectric permeability of the material  $\varepsilon$  in the range from 3.56 to 12.20. It was also found that the increase in the share of  $\text{SrAl}_2\text{Si}_2\text{O}_8$  leads to a significant reduction in the shrinkage of ceramics to almost a zero value. The results reported in [13, 14] indicate the feasibility of receiving polyphase radio transparent ceramics at a reduced firing temperature, as well as demonstrate the possibility to adjust their properties by varying the phase composition, which is certainly an advantage.

Our analysis of existing developments in a given field of materials science has revealed that the main drawbacks of glass crystalline and ceramic RTM are the constraints for the temperature of operation and the energy intensity of manufacturing. The reproducibility of material properties has also remained unresolved. The reason for this may be the difficulties associated with the empirical approach to the RTM design, the fundamental impossibility to create a material that could meet the entire set of requirements, the application of a "trial and error" method. The absence of an effective methodology for the RTM design entails large material, time, and energy costs, which render such studies impractical.

A variant of overcoming related difficulties may be a scientifically substantiated approach to the design of materials with predetermined properties, based on the analysis of state diagrams of the basic phase-forming oxides. This would avoid miscalculations in selecting the synthesis conditions of target phases that could ensure the required properties, and minimize the cost of designing new functional materials. This approach was applied in works [15, 16] to create functional materials and coatings by the targeted synthesis of a set of phases based on the study of the subsolidus structure of basic systems. A prerequisite for the implementation of a given technique to design a material with specified properties is the presence of reliable data about the structure of the basic oxide system, which are generalized in the diagram of its state. The state diagram of an oxide system gives an idea of the phase equilibrium, stability conditions, melting temperatures, and polymorphic phase transformations, the existence of solid solutions, as well as other information necessary to create materials with predefined properties.

Underlying the creation of polyphase ceramic and glass crystalline materials are mainly the multi-component oxide systems. Acquiring experimental data describing phase equilibrium in multicomponent systems requires enormous material and time costs. For example, it takes an average of 5 years to study each of the four triple subsystems [17]. Given the above, we have accepted such an alternative to establishing co-existing phases in multicomponent oxide systems as the application of comprehensive thermodynamic calculations whose implementation is possible in the presence of the compounds' thermodynamic properties. The data that would be acquired by using this method could be used as a starting point for selective experiments in the studies by materials scientists, which, in general, might contribute to accelerating the innovations when designing new materials [18].

The relevance of phase equilibrium studies in the ZnO–SrO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> system is predetermined by the increased demand by various industries for products made from refractory non-metallic and silicate materials derived on the basis of this system's compounds. In the global market, this trend is due to the growing demand for special-purpose products for engineering, energy, microelectronics, and medicine, including made from sitalls and ceramic structural materials, which are combined by a given system.

The interest in the  $ZnO-SrO-Al_2O_3-SiO_2$  system in terms of aviation and rocket industries is due to the fact that it includes compounds with the required level of dielectric characteristics, which is important in the design of radio-transparent materials. A series of compounds within the system are characterized by a high melting point and a relatively small temperature expansion and, therefore, can provide the materials with heat resistance and thermal stability (Table 1).

#### Table 1

Properties of compounds from the ZnO–SrO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> system that are of interest to synthesize heat-resistant radio transparent materials (according to data from) [19]

Compound	Density p, kg/m³	Melting point <i>T</i> , °C	CLTE, α·10 <sup>6</sup> 1/K	Dielectric charac- teristics*		
				ε	tgδ·10 <sup>-4</sup>	
Quartz SiO <sub>2</sub>	2,648	1,713	0.5	3.8	0.5 - 60.0	
Corundum	-	2,050	2.65	6.5	2.0-5.0	
Willemite Zn <sub>2</sub> SiO <sub>4</sub>	4,000	1,512	3.2	5.5	0.3–0.8	
Strontium anorthite SrAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub>	3,270	1,654	6.5	6.4	11.0–50.0	
$\begin{array}{c} Gahnite\\ Al_2ZnO_4 \end{array}$	3,600	1,950	6.5	8.5	4.0-6.0	
Mullite Al <sub>6</sub> Si <sub>2</sub> O <sub>13</sub>	3,100	1,830	6.2	7.0	5.0-10.0	
*at a frequency of 1 MHz at 20 °C						

The compounds in Table 1 possess a set of all required properties and are potentially suitable for the synthesis of radio transparent ceramics. A comparative analysis of the physical-mechanical, thermal-physical, and dielectric properties of these compounds reveals that it is appropriate to obtain radio-transparent ceramics with a lower synthesis temperature based on the combinations of phases: "willemite+quartz", "willemite+strontium anorthite".

It should be noted that at present there are no data on the systemic experimental study of phase equilibria in the ZnO-SrO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system required to build a state diagram. Only binary subsystems, as well as two triple subsystems ZnO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> and SrO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>, are described fully enough [20]. There are no state diagrams for the triple subsystems SrO-ZnO-SiO<sub>2</sub> and SrO-ZnO-Al<sub>2</sub>O<sub>3</sub>.

### 3. The aim and objectives of the study

The aim of this study is to examine the subsolidus structure of the  $SrO-ZnO-Al_2O_3-SiO_2$  system and to substantiate the region of oxide compositions to produce heat-resistant radio-transparent ceramics. This would make it possible to rationally design materials' compositions taking into consideration the specified properties and operational conditions, as well as improve the reproducibility of the ceramics' phase composition and properties.

To achieve the set aim, the following tasks have been solved:

– to conduct a geometric-topological analysis of the  $SrO-ZnO-Al_2O_3$ –  $SiO_2$  system, including the identification of combinations of coexisting phases and the calculations of the geometric-topological characteristics of elementary tetrahedra;

– to substantiate the selection of the region of oxide compositions of the specified system, using which could enable the synthesis of phases ( $Zn_2SiO_4$  and  $SrAl_2Si_2O_8$ ); and to develop the compositions of masses with a different ratio of target phases;

- to investigate the functional properties and to determine the structural-phase features of the new materials and to draw a conclusion about their applicability.

## 4. Materials and methods to study the subsolidus state of a four-component system and the properties of ceramics

The subsolidus state of the ZnO – SrO –  $Al_2O_3$  – SiO<sub>2</sub> system was investigated using a geometric-topological analysis whose methodology is detailed in work [21]. A given method involves the use of barycentric coordinates and elements from Euclidean geometry. Given this, the conode lengths of elementary tetrahedra were calculated from the following formula

$$L^{2} = (X_{2} - X_{1})^{2} + (Y_{2} - Y_{1})^{2} + (Z_{2} - Z_{1})^{2} + (X_{2} - X_{1})(Y_{2} - Y_{1}) + (X_{2} - X_{1})(Z_{2} - Z_{1}),$$
(1)

where  $x_1$ ,  $y_1$ ,  $z_1$ ,  $x_2$ ,  $y_2$ ,  $z_2$  are the coordinates of the coexisting phase pairs (the concentration of components in % by weight). The volumes of elementary tetrahedra  $(V_i)$  were determined by finding a determinant taking into consideration the barycentric coordinates

$$V_{i} = \begin{vmatrix} X_{1} & Y_{1} & Z_{1} & 1 \\ X_{2} & Y_{2} & Z_{2} & 2 \\ X_{3} & Y_{3} & Z_{3} & 3 \\ X_{4} & Y_{4} & Z_{4} & 4 \end{vmatrix},$$
(2)

where  $X_i$ ,  $Y_i$ ,  $Z_i$ ,  $T_i$  is the content of oxides in the compounds that make up the elementary tetrahedron.

The degree of asymmetry  $(K_a)$  of the elementary tetrahedra was determined as a ratio of the maximum  $(L_{max})$  and minimum  $(L_{min})$  edge lengths

$$K_a = L_{\max} / L_{\min}.$$
 (3)

Given that at the point of eutectics the temperature of the liquidus curves for all components of the system are the same, we calculated the temperature and composition of eutectics for a four-component system by solving the following system of equations

$$T_{1} = T_{\text{melt},1} / (1 - \ln(X_{1}) / N_{1}) =$$

$$= T_{2} = T_{\text{melt},2} / (1 - \ln(X_{2}) / N_{2}),$$

$$T_{2} = T_{\text{melt},2} / (1 - \ln(X_{2}) / N_{2}) =$$

$$= T_{3} = T_{\text{melt},3} / (1 - \ln(X_{3}) / N_{3}),$$

$$T_{3} = T_{\text{melt},3} / (1 - \ln(X_{3}) / N_{3}) =$$

$$= T_{4} = T_{\text{melt},4} / (1 - \ln(X_{4}) / N_{4}),$$

$$X_{1} + X_{2} + X_{3} + X_{4} = 1.$$
(4)

For the preparation of samples, we used quartz sand with a SiO<sub>2</sub> content of 99.8 % by weight, alumina G-00, strontium carbonate, and white zinc paint with a ZnO content of 99.9 % by weight. The raw mixtures were obtained by joint dry grinding in a ball mill over 18 hours. The samples were molded by a semi-dry pressing using a 15 % solution of dextrin as a temporary ligament. The samples were dried in a drying chamber at 110 °C to a residual humidity of no more than 1 %. The samples were fired in a muffle furnace at 1,200 °C. The aging duration at the maximal temperature was 1 hour.

The following basic characteristics of baking were determined for the resulting samples of ceramics by a method of hydrostatic weighing in water: linear shrinkage (*L*, %), bulk density ( $\rho_{BD}$ ), water absorption (*W*) and open porosity ( $P_{open}$ ). The compressive load was increased evenly at such a rate that the time period from applying the load to the sample destruction lasted from 20 to 40 s. The error of measuring a destructive force did not exceed 1 %. We determined the strength limit at bending at the laboratory instrument MII-100N for the samples in the form of tiles, the size of  $15 \times 5 \times 80$  mm. The strength limit of samples at bending was calculated on the basis of the value of the destructive effort at a three-point bending.

To determine the permittivity and the tangent of a dielectric loss angle, we used samples in the form of tablets, 20 mm in diameter and 3 mm in height. The measurements were performed at the automated device RLC E7-8 (Ukraine), designed to measure the characteristics of dielectric losses in the range of 0.0001-1.0 at a voltage range of 0-270 V.

The phase composition of the examined samples was determined by X-ray phase analysis (XPA) using the diffractometer DRON-3M (RF) with a CuK $\alpha$ -radiation and a nickel filter under the standard conditions of its operation. The American ASTM database was used to identify the phases.

We studied the microstructure of samples by a method of electron-microscopic analysis, using the scanning electron microscope JSM-6390LV (Japan) under the mode of secondary electrons.

# 5. Studying the subsolidus structure of the ZnO–SrO– $Al_2O_3$ –SiO<sub>2</sub> system

The ZnO–SrO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> system, with the coexistence of the phases of strontium anorthite  $SrAl_2Si_2O_8$  and willemite  $Zn_2SiO_4$  in some of its sections, was chosen as the base for synthesizing heat-resistant ceramic RTM. The most interesting are the regions of oxide compositions, which allow the simultaneous synthesis of target phases.

As part of our study of the subsolidus structure of the ZnO–SrO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> system, we determined the geometric-topological characteristics of the elementary tetrahedra into which the internal volume of a four-component system was split, according to data by *Materials Project* (Fig. 1) [22].



Geometric-topological characteristics such as elementary volume ( $_{\Delta}V$ ,  $_{\infty}$ ) and degree of asymmetry ( $K_a$ ) make it possible to assess the manufacturability of compositions of raw mixtures designed by using the system's oxide compositions. The maximum values of  $_{\Delta}V$  and the minimum values of  $K_a$  are preferred in terms of the accuracy of the dosage of the components of technological mixtures, as well as the minimum mixing time required for averaging the components. In turn, the compositions and temperatures of the eutectics of tetrahedra give an idea of the minimum temperature of synthesis and the phase composition of materials derived from them. In addition, these data make it possible to predict the temperature of the material's operation.

The study results have shown that the ZnO–SrO–Al<sub>2</sub>O<sub>3</sub>– SiO<sub>2</sub> system is split into 20 elementary tetrahedra:

1) $Al_2O_3$ -ZnO· $Al_2O_3$ -SrO· $Al_2O_3$ ·2SiO<sub>2</sub>-3 $Al_2O_3$ ·2SiO<sub>2</sub>;

2) 
$$Al_2O_3$$
-ZnO·Al\_2O\_3-SrO·Al\_2O\_3·2SiO\_2-SrO·2Al\_2O\_3;

3) ZnO·Al<sub>2</sub>O<sub>3</sub>-SrO·Al<sub>2</sub>O<sub>3</sub>-2SrO·Al<sub>2</sub>O<sub>3</sub>·SiO<sub>2</sub>-SrO× ×2Al<sub>2</sub>O<sub>3</sub>;

5) SrO–SrO·ZnO–3SrO·Al<sub>2</sub>O<sub>3</sub>–2SrO·SiO<sub>2</sub>;

6) SiO<sub>2</sub>-3Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>-ZnO·Al<sub>2</sub>O<sub>3</sub>-SrO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>;

7) ZnO-2SrO·Al<sub>2</sub>O<sub>3</sub>·SiO<sub>2</sub>-SrO·Al<sub>2</sub>O<sub>3</sub>-ZnO·Al<sub>2</sub>O<sub>3</sub>;

8)  $2\text{SrO} \cdot 2\text{SiO}_2 - 2\text{SrO} \cdot \text{SiO}_2 - 2\text{SrO} \cdot \text{Al}_2\text{O}_3 \times$ 

 $\times$ SiO<sub>2</sub>;

9) 2SrO·ZnO·2SiO<sub>2</sub>–2SrO·Al<sub>2</sub>O<sub>3</sub>·SiO<sub>2</sub>–SrO·Al<sub>2</sub>O<sub>3</sub>× ×2SiO<sub>2</sub>–SrO·SiO<sub>2</sub>;

10)  $SiO_2$ -2 $SrO\cdot ZnO\cdot 2SiO_2$ -SrO·Al<sub>2</sub>O<sub>3</sub>·2 $SiO_2$ -SrO·SiO<sub>2</sub>;

11)  $ZnO-SrO\cdotAl_2O_3-2SrO\cdotSiO_2-3SrO\cdotAl_2O_3;$ 

12)  $ZnO-SrO\cdotAl_2O_3-2SrO\cdotAl_2O_3\cdotSiO_2-2SrO\cdotSiO_2;$ 

13) ZnO·Al<sub>2</sub>O<sub>3</sub>-2SrO·ZnO·2SiO<sub>2</sub>-2SrO·Al<sub>2</sub>O<sub>3</sub>× ×SiO<sub>2</sub>-SrO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>;

14)  $SiO_2$ -2SrO·ZnO·2SiO<sub>2</sub>-2ZnO·SiO<sub>2</sub>-SrO·Al<sub>2</sub>O<sub>3</sub>× ×2SiO<sub>2</sub>;

15)  $ZnO \cdot Al_2O_3 - 2SrO \cdot ZnO \cdot 2SiO_2 - SrO \cdot Al_2O_3 \times \times 2SiO_2 - 2ZnO \cdot SiO_2;$ 

16) SiO<sub>2</sub>–ZnO·Al<sub>2</sub>O<sub>3</sub>–2ZnO·SiO<sub>2</sub>–SrO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>;

17)  $ZnO-2SrO\cdot ZnO\cdot 2SiO_2-2SrO\cdot Al_2O_3\cdot SiO_2-2SrO \times \times SiO_2;$ 

18) ZnO–2SrO·ZnO·2SiO<sub>2</sub>–ZnO·Al<sub>2</sub>O<sub>3</sub>–SrO·Al<sub>2</sub>O<sub>3</sub>·SiO<sub>2</sub>; 19) ZnO–SrO·ZnO–2SrO·SiO<sub>2</sub>–3SrO·Al<sub>2</sub>O<sub>3</sub>;

20)  $ZnO-2ZnO\cdotSiO_2-2SrO\cdotZnO\cdot2SiO_2-ZnO\cdotAl_2O_3$ .

The basic geometric-topological characteristics of the tetrahedra, which determine the subsolidus state of the examined system, are given in Table 2.

An analysis of data in Table 2 suggests that there are several large tetrahedra within the  $ZnO-SrO-Al_2O_3-SiO_2$  system where there are phases of willemite  $Zn_2SiO_4$  or strontium anorthite  $SrAl_2Si_2O_8$ :

SiO<sub>2</sub>-Sr<sub>2</sub>ZnSi<sub>2</sub>O<sub>7</sub>-Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> ( $\Delta V$ =70.37‰;  $K_a$ =2.56);

SiO<sub>2</sub>-Sr<sub>2</sub>ZnSi<sub>2</sub>O<sub>7</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>-SrSiO<sub>3</sub> (
$$_{\Delta}V$$
=78.85‰;  $K_a$ =2.2);

SiO<sub>2</sub>-3Al<sub>6</sub>Si<sub>2</sub>O<sub>13</sub>-ZnAl<sub>2</sub>O<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> ( $\Delta V$ =88.81‰;  $K_a$ =2.74);

SiO<sub>2</sub>-ZnAl<sub>2</sub>O<sub>4</sub>-Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> ( $\Delta V$ =129.07‰;  $K_a$ =1.8).

At the same time, only in two elementary tetrahedra (SiO<sub>2</sub>-Sr<sub>2</sub>ZnSi<sub>2</sub>O<sub>7</sub>-Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> and SiO<sub>2</sub>-ZnAl<sub>2</sub>O<sub>4</sub>-Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>) the phases of Zn<sub>2</sub>SiO<sub>4</sub> and SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> coexist. This indicates the possibility of simultaneous synthesis of a combination of target phases when using oxide compositions belonging to these tetrahedra.

It should be noted that the tetrahedra, which include gahnite, corundum, mullite, and strontium anorthite, are also suitable for the synthesis of RTM; however, they are characterized by higher eutectic temperatures. That, on the one hand, could make it possible to increase the maximum temperature of operation but, on the other hand, would significantly elevate the synthesis temperature of polyphase ceramics.

Table 2

Totrahodron	Totrohodron volume V %	Asymmetry degree $K_a$	Futaatias tomporeture T V	Eutectics composition, %			
Tetraneuron	Tetraneuron volume $\Delta V$ , 70		Eutectics temperature <i>I<sub>e</sub></i> , K	1	2	3	4
1	52.83	1.88	1,788.60	23.74	18.26	36.56	21.43
2	55.21	1.76	1,804.50	25.13	19.73	41.39	13.74
3	12.09	3.33	1,815.90	20.82	16.20	47.98	14.99
4	38.07	2.81	1,631.72	7.91	79.45	9.51	3.12
5	24.45	1.95	1,455.63	15.68	83.10	0.81	0.41
6	88.81	2.74	1,647.68	66.35	9.48	8.68	15.49
7	45.68	4.38	1,795.46	18.88	29.72	38.58	12.80
8	15.60	2.7	1,583.36	54.68	0.59	34.66	10.07
9	25.50	1.81	1,572.92	50.29	4.58	5.36	39.77
10	78.85	2.2	1,507.65	37.50	29.04	2.69	30.77
11	56.02	4.01	1,809.67	61.67	15.72	4.74	17.87
12	31.35	4.59	1,771.14	58.43	12.97	24.79	3.81
13	48.20	2.6	1,625.64	7.64	75.90	7.48	8.98
14	70.37	2.56	1,465.00	57.20	19.59	21.56	1.64
15	38.74	1.48	1,572.76	5.53	50.23	5.35	38.88
16	129.07	1.8	1,522.00	62.05	3.98	29.82	3.14
17	43.76	3.62	1,575.90	42.67	51.54	4.72	1.07
18	63.79	2.19	1,567.00	42.04	48.23	5.36	4.36
19	31.12	3.84	1,397.03	29.62	69.72	0.24	0.42
20	46.24	2.26	1,512.00	37.83	28.26	30.18	3.72

Characteristics of the elementary tetrahedra of the  $ZnO-SrO-Al_2O_3-SiO_2$  system

# 6. Substantiation of the region of oxide compositions for designing heat-resistant radio transparent ceramics

The results of studying the phase equilibria in the subsolidus region of the ZnO-SrO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system make it possible to predict the phase composition of materials both at the stage of RTM synthesis and when operating the fairings. Our analysis of the geometric-topological characteristics of the tetrahedra that include the target phases revealed that it is preferable to use a  $SiO_2$ -ZnAl<sub>2</sub>O<sub>4</sub>-Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> tetrahedron to design RTM as a less asymmetrical ( $K_a$ =1.8) and possessing a large elementary volume ( $_{\Lambda}V=129.07$  %). From a manufacturing point of view, the composition of a given tetrahedron could ensure the reproducibility of the phase composition at the existing industrial dosage accuracy (0.1 g) and would reduce the time for averaging a technological mixture. Given the fact that the eutectic temperature of this SiO<sub>2</sub>-ZnAl<sub>2</sub>O<sub>4</sub>-Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> tetrahedra is 1,522 K, it can be assumed that the lower temperature limit of the material operation would be at least 1,200 °C. The analysis of the eutectic composition (in molar %:  $SiO_2 - 62.05$ ;  $ZnAl_2O_4 - 3.98$ ;  $Zn_2SiO_4 - 29.89$ ;  $SrAl_2Si_2O_8 - 3.14$ ) indicates the need to select oxide compositions in the region adjacent to the edge Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>.

If it is necessary to increase the working temperatures of fairings (>1,200 °C), the compositions of a given tetrahedron make it possible to adjust the phase composition of ceramics by increasing the proportion of gahnite, as a phase with a high melting point (T=1,950 °C).

## 7. The formulations and properties of radio transparent ceramics designed on the basis of the system's compositions

We designed the compositions to produce radio-transparent ceramics within the concentrations of the elementary tetrahedra  $SiO_2-ZnAl_2O_4-Zn_2SiO_4-SrAl_2Si_2O_8$ . To study the effect of the ratio of the target phases ( $SrAl_2Si_2O_8$  and  $ZnSiO_4$ ) on the ceramics' properties, we investigated three compositions whose ratio of the specified phases varied as 1:3; 1:1; 3:1. The properties of the ceramic materials fired at 1,200 °C are given in Table 3.

Table 3

Dava setas	The properties of samples with the following ratio of the target phases SrAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub> :ZnSiO <sub>4</sub>				
Property	P-1	P-2	P-3		
	1:3	1:1	3:1		
Water absorption W, %	16.87	22.54	21.45		
Open porosity Popen, %	39.18	44.86	42.68		
Bulk density $\rho_{BD}$ , kg/m <sup>3</sup>	2080	1980	1990		
Linear shrinkage L, %	3.31	5.29	1.02		
Strength limit at compression $\sigma_c$ , MPa	16.79	21.53	19.10		
Strength limit at flexure $\sigma_f$ , MPa	14.04	11.17	9.88		
Permittivity, $\epsilon^*$	8.62	5.98	7.96		
Dielectric loss tangent, tgδ*	0.008	0.004	0.007		
*at a frequency of 1 MHz					

Our results of studying the properties of the obtained ceramic materials show that the ceramics' samples meet the requirements for RTM in terms of the level of dielectric characteristics. The sample of composition P-2, whose assigned phase ratio SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>:ZnSiO<sub>4</sub>=1:1, is characterized by the minimal indicators ( $\varepsilon$ =5.98; tg $\delta$ =0.004 at a 1 MHz frequency). When the SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> share in the P-3 sample increases, there is a significant decrease in shrinkage at firing. The resulting materials are characterized by a low strength at compression and bending, which is due to their high porosity. In order to improve the strength indicators of the resulting RMP, a higher level of caking is needed while maintaining their phase composition.

The structural-phase features of the obtained radio transparent ceramics were investigated by XPA methods and scanning electron microscopy. The results from a qualitative X-ray phase analysis of the samples are shown in Fig. 2. As evidenced by the data, the materials obtained differ in the quantitative and qualitative content of crystalline new formations. The sample of P-1 composition, whose assigned phase ratio of SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>:ZnSiO<sub>4</sub> is 1:3, demonstrates by its radiograph (Fig. 2, a) the largest number of reflexes of Zn<sub>2</sub>SiO<sub>4</sub> willemite at maximum intensity (d=0.699; 0.403; 0.349; 0.2837; 0.2636; 0.2318 nm). That allows us to conclude that this phase dominates in the material. The strontium anorthite SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> phase is characterized by much less intensity of reflexes. However, its presence is doubted given the large number of peaks on the radiograph (d=0.457; 0.376; 0.345; 0.327; 0.322; 0.2935; 0.2552 nm). The zincite ZnO phase was detected in the sample in an impurity amount (d=0.282; 0.2605; 0.2479; 0.1914 nm). For the P-2 sample, whose assigned phase ratio is  $SrAl_2Si_2O_8$ :ZnSiO<sub>4</sub>=1:1, the number and intensity of the reflexes of willemite and strontium anorthic on the radiograph (Fig. 2, b) are almost the same, which indicates an approximately equal number of these phases in the material.

The concomitant phases in the P-2 sample were not identified, indicating the completeness of the course of the reaction that forms the target phases. The radiograph of sample P-3 (Fig. 2, c), whose assigned phase ratio is  $SrAl_2Si_2O_8:ZnSiO_4=$ =1:3, is dominated, by the number and intensity, by the reflexes of strontium anorthite; willemite is present but in a smaller amount. The concomitant phases in a given material were not identified either. Thus, the results from XPA

explain the established indicators of the dielectric properties of samples with a different ratio of target phases and make it possible to choose the optimal composition of ceramics P-2 with the  $SrAl_2Si_2O_8$ :ZnSiO<sub>4</sub>= 1:1 ratio.

As demonstrated by the results from electron microscopy (Fig. 3), the structure of the ceramic sample is fine crystalline, homogeneous, without defects and cracks.

The image with a magnification of  $\times 300$  (Fig. 3, *a*) makes it possible to assess the natural roughness of the chipped surface, the sample porosity, and the homogeneity of the material's structure. SEM images with a magnification of ( $\times 5,000$ ) (Fig. 3, *b*, *c*) show separate areas of the ceramics. Section 1 (Fig. 3, *c*) demonstrates the clusters of short-prismatic willemite crystals measuring 4–5 µm. Over area 2 (Fig. 3, *c*), we registered the new formations of monoclinic strontium anorthic, whose size amounts to 25–30 µm. The prism-shaped crystals are formed by layers oriented along the basic crystallographic axis [010] (Fig. 3, *c*). The morphological features of SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> partly explain the small shrinkage and high porosity of the ceramics whose phase composition is dominated by this compound.



Fig. 2. Radiographs of the samples fired at a temperature of 1,200 °C: a – sample P-1; b – sample P-2; c – sample P-3;  $\Rightarrow$  – Zn<sub>2</sub>SiO<sub>4</sub>;  $\Rightarrow$  – SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>;  $\equiv$  – ZnO







# 8. Discussion of studying the structure of the ZnO-SrO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system and the designed radio transparent ceramics

A comparative analysis of the  $ZnO-SrO-Al_2O_3-SiO_2$  system's compounds reveals the prospect of obtaining the heat-resistant radio-transparent ceramics based on the phases of willemite and strontium anorthite, which possess satisfactory dielectric characteristics (Table 1). This choice is due to the need to reduce the energy intensity in the production of fairings and other special-purpose structures. This task can be solved by reducing the synthesis temperature of the target phases, which would provide for the functional properties of the material. In contrast to the empirical approach to the selection of ceramic composition, our study has employed a composition design methodology based on an analysis of the subsolidus state of the system, which includes the basic phase-forming oxides.

Underlying the creation of radio-transparent ceramics with a

predefined phase composition are the results of studying the subsolidus structure of the basic oxide system  $ZnO-SrO-Al_2O_3-SiO_2$  (Fig. 1, Table 2). To ensure the synthesis of a combination of phases "willemite+strontium anorthite" at a lower temperature (~1,200 °C), we have selected the region of oxide compositions within the concentrations of the elementary tetrahedron SiO<sub>2</sub>-ZnAl<sub>2</sub>O<sub>4</sub>- $Zn_2SiO_4$ -SrAl\_2Si\_2O\_8 (Fig. 1). The geometric-topological characteristics of a given tetrahedron ( $_{\Delta}V=129.07$  ‰,  $K_a=1.8$ ) indicate the possibility for obtaining radio-transparent ceramics with stable dielectric characteristics at a lower synthesis temperature. Taking into consideration the requirements for the accuracy of dosage and preparation of the technological mixture, the oxide compositions of a given tetrahedron are easier to manufacture as they warrant the reproducibility of the phase composition. In order to produce radio-transparent ceramics at a reduced synthesis temperature of the target phases of Zn<sub>2</sub>SiO<sub>4</sub> and SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>, one must select compositions in the region adjacent to the Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> edge of the tetrahedron. At the same time, the lower limit of working temperatures is projected at 1,200 °C. To obtain ceramics with an operational temperature exceeding 1,200 °C, the oxide compositions must be located at the ZnAl<sub>2</sub>O<sub>4</sub>-Zn<sub>2</sub>SiO<sub>4</sub>-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> edge of the tetrahedron, which would make it possible to complement the phase composition with gahnite  $ZnAl_2O_4$ , whose melting point is high (Table 1).

The results from experimental studies of samples whose compositions belong to the SiO2-ZnAl2O4-Zn2SiO4-SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> tetrahedron show that it is possible to obtain the polyphase ceramics at 1,200 °C, whose dielectric properties could meet the requirements for RTM. The phase composition of the materials obtained is characterized by the presence of two target phases ( $Zn_2SiO_4$  and  $SrAl_2Si_2O_8$ ), the amount of which correlates with their ratio assigned when designing the compositions. The presence of the concomitant zincite phase, identified in the P-1 sample (Fig. 2, a), indicates an excess amount of zinc oxide in the raw mixture. It is noteworthy that the indicators of permittivity for zincite ( $\varepsilon$ =8.5) meet the requirements for RTM ( $\epsilon$ =1÷10). However, the presence of ZnO increases the P-1 sample indicator  $\varepsilon$  compared to the samples of P-2 and P-3 (Table 3), whose basic phases are  $SrAl_2Si_2O_8$  ( $\epsilon$ =6.4) and  $ZnSiO_4$  ( $\epsilon$ =5.5). The relatively high porosity of the ceramics is due to both the insufficient degree of baking and the layered structure of the strontium anorthite's new formations.

The disadvantage of the materials obtained is insufficient strength (Table 3), which can be improved by increasing the degree of ceramic baking while maintaining the target phase composition. To solve this task, it seems promising to introduce additives in small amounts (up to 2 % by weight above 100 %) that would act as the intensifiers of baking and phase formation, Such additives could be  $Li_2O$  or the eutectic composition of the  $Li_2O$ -SnO<sub>2</sub> system whose baking effectiveness was proven in work [23]. This technique could reduce the porosity and increase the strength of ceramics without increasing the temperature of article firing, which, in turn, would ensure maintaining the specified phase composition of materials.

### 9. Conclusions

1. The results of our study have established the subsolidus structure of the ZnO–SrO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> system, which is of interest when forming heat-resistant radio-transparent ceramic materials. For 20 elementary tetrahedra of the system, the basic geometric-topological characteristics have been defined: the elementary volumes, the degrees of asymmetry, the temperature and compositions of eutectics. Data about the phase ratios of the ZnO–SrO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> system is of interest when designing functional ceramics, protective coatings, special types of cement, and luminophores.

2. We have selected, within the concentrations of the elementary  $SiO_2-ZnAl_2O_4-Zn_2SiO_4-SrAl_2Si_2O_8$  tetrahedron, the region of oxide compositions that ensure the stability of the RTM dielectric characteristics, the reproducibility of properties, and the manufacturability of compositions. In order to produce polyphase radio transparent ceramics with a reduced synthesis temperature for the combinations of "willemite+strontium anorthite" phases, the compositions must be located near the edge of the  $Zn_2SiO_4-SrAl_2Si_2O_8$  tetrahedron. The design of RTM with an operating temperature exceeding 1,200 °C is possible when the phase composition is adjusted by forming a high-temperature phase of gahnite  $ZnAl_2O_4$ .

3. We have experimentally obtained ceramic materials containing only a specified combination of phases, which confirms the effectiveness of the methodology of designing the compositions of functional materials based on the analysis of phase equilibria in the system of phase-forming oxides. In terms of the level of dielectric characteristics, the designed ceramics meet the requirements for RTM ( $\epsilon$ =5.68–8.68; tg $\delta$ =0.004–0.008). It has been shown that it is possible to adjust the properties of a ceramic material when the ratio of the target phases Zn<sub>2</sub>SiO<sub>4</sub> and SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> is changed from 1:3 to 3:1 by varying the phase-forming oxides. We have established the optimal phase ratio of Zn<sub>2</sub>SiO<sub>4</sub>:SrAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>=1:1, providing for the ceramics with low dielectric permeability ( $\varepsilon$ =5.98) and minimal losses (tg $\delta$ =0.004). In the future, the strength of the materials can be increased by the introduction of baking intensifying additives that would not participate in the processes of phase formation, which could maintain the specified phase composition and the material's properties. Resolving this task in general could improve the resource consumption and reliability of antenna fairings, as well as improve the handling of aircraft.

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