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RADIO-TRANSPARENT CERAMICS IN THE $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ SYSTEM: PHYSICAL, MECHANICAL, THERMAL AND DIELECTRIC PROPERTIES

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The production of phase shifters for phased-array antennas is typically based on cordierite glass-ceramics, such as grades ST-32 and ST-38, or on materials of similar composition. Such materials are required to exhibit water resistance, low dielectric losses in the microwave frequency range, controlled values of the coefficient of thermal expansion, and high thermomechanical performance. This study focuses on the development of low-temperature ceramic compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system for the fabrication of phase shifters with tailored physical, mechanical, thermal, and dielectric properties. Thermodynamic calculations were performed to evaluate possible reactions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system involving components of eutectic glass compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ system. The triple eutectic point of this system was considered, at which the following phases crystallized: cordierite ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$), forsterite ($2\text{MgO}\cdot \text{SiO}_2$), and clinoenstatite ($\text{MgO}\cdot \text{SiO}_2$). The thermodynamic analysis included the determination of Gibbs free energy changes for possible reactions leading to the formation of cordierite and forsterite phases, as well as reactions involving TiO_2 . A relationship between the physical, mechanical, and thermal properties of the developed ceramics and the technological parameters of their processing (sintering temperature and TiO_2 content) was established. Machine learning methods made it possible to quantitatively assess the relative contributions of the controlled factors and to improve the prediction of the properties of the developed ceramics within the investigated parameter range. The most rational compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system were identified, providing a favorable combination of properties and enabling sintering at 1200°C.

Keywords: radio-transparent ceramics, phase shifters, cordierite, eutectic glass, titanium dioxide, thermodynamic analysis, sintering, physical and mechanical properties.

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Introduction

The development of advanced technologies in aerospace and rocket engineering continuously requires the design and implementation of a wide range of

dielectric materials, including glass-ceramic and ceramic systems [1]. These materials must meet stringent requirements in terms of dielectric, thermal, and mechanical properties. Glass-ceramic and ceramic

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materials based on aluminosilicate systems satisfy these requirements and are relatively cost-effective [2]. The required functional characteristics can be achieved through the use of ternary compounds such as spodumene ($\text{Li}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot 4\text{SiO}_2$), eucryptite ($\text{Li}_2\text{O}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$) [3], cordierite ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$) [4], celsian ($\text{BaO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$) [5] and strontium anorthite ($\text{SrO}\cdot\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$) [6,7].

Among alkali-free aluminosilicate materials, cordierite is of particular interest owing to its favorable combination of high thermomechanical performance, low density, and low dielectric losses in the microwave frequency range [8]. Cordierite-based materials are traditionally used in the fabrication of radio-transparent components, including phase shifters, which are essential elements of phased-array antenna systems for radar applications.

Materials for phase shifters must exhibit low dielectric losses in the microwave frequency range, a relative permittivity (ϵ) within a specified range, and prescribed values of the coefficient of thermal expansion (CTE) [9]. Phase shifters are typically produced from cordierite glass-ceramics such as ST-32 and ST-38 or compositionally similar materials. These grades are characterized by CTE values of approximately $32\cdot 10^{-7} \text{ }^\circ\text{C}^{-1}$ and $38\cdot 10^{-7} \text{ }^\circ\text{C}^{-1}$, respectively.

Glass-ceramic materials in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ (MAS) system, obtained by glass-based or ceramic (powder-based) processing routes, require high melting temperatures of the initial glass (1550–1600°C). In addition, conventional glass and powder technologies for producing cordierite-based glass-ceramics involve several challenges, particularly in selecting optimal glass compositions and efficient heat-treatment regimes.

The active formation of the cordierite phase in ceramics derived from natural raw materials (kaolinite, talc, and crystalline alumina) occurs within the temperature range of 1160–1270°C. However, even sintering at temperatures of 1400–1450°C for extended durations (20–60 h) does not yield a dense material with minimal water absorption (<0.1%). Moreover, the formation of significant amounts of secondary phases such as spinel, mullite, and clinoenstatite adversely affects the performance characteristics of such ceramics [10].

This study aims to develop low-temperature ceramic compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system for the fabrication of phase shifters as analogs of ST-32 cordierite glass-ceramic, with controlled physical, mechanical, thermal, and dielectric properties.

Materials and methods

Thermodynamic calculations were performed to evaluate the possible reactions in the $\text{MgO}-\text{Al}_2\text{O}_3-$

$\text{SiO}_2-\text{TiO}_2$ system involving components of eutectic glass compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ (MAS) system. The triple eutectic point of the MAS system was considered, at which the following phases crystallize cordierite ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$), forsterite ($2\text{MgO}\cdot\text{SiO}_2$), and clinoenstatite ($\text{MgO}\cdot\text{SiO}_2$). The thermodynamic analysis included the determination of the Gibbs free energy changes for possible reactions leading to the formation of cordierite and forsterite phases involving the eutectic-glass components of the MAS system, as well as reactions involving TiO_2 . Metakaolin ($\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$), formed by the dihydroxylation of kaolinite, was considered as one of the initial components in the analyzed reactions. Kaolin, whose principal mineral phase is kaolinite, was used in ceramic compositions as a suspending additive, making it possible to fabricate products of varying shape complexity.

The phase composition of the ceramics was analyzed by X-ray diffraction (XRD) using a DRON-3 diffractometer with Co-K α radiation.

The raw materials used for the preparation of the experimental ceramics included eutectic MAS glass, enriched kaolin (grade ZREF-1, Ukraine), technical alumina (grade G-0), magnesium hydroxide (analytical grade), silicon dioxide (grade A), and titanium dioxide (analytical grade). Ceramic slips with a moisture content of 26–27 wt.% were prepared by joint wet milling of the raw components. The samples were cast into plaster molds in the form of cylinders, bars, and disks, followed by drying to a residual moisture content below 1%. Sintering was carried out in an air atmosphere using an SNOL 15/1300 electric furnace at temperatures of 1100–1250°C.

For the preparation of eutectic MAS glass, technical alumina (grade G-0), silicon dioxide (grade A), and boric acid (analytical grade) were used. The addition of B_2O_3 (10 weight parts over 100 wt.%) reduced the glass-melting temperature without changing the mineralogical composition of the crystalline phases in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3-\text{SiO}_2$ (MABS) system. The MABS glass was melted in an electric furnace with silicon carbide heaters at 1375°C for 1 h using corundum crucibles.

Water absorption (W), open porosity (P), and apparent density (ρ) of the ceramic samples were determined by the saturation method followed by weighing in air and in water.

Compressive strength (σ) of cylindrical samples ($D=H=10$ mm) was measured using a PSU-10 hydraulic press.

The coefficient of thermal expansion (CTE) was determined using an automatic quartz dilatometer

(DKV-5A) in the temperature range of 20–400°C on samples of 5×5×50 mm.

Thermal shock resistance was evaluated based on the maximum temperature difference (ΔT , °C) sustained by the samples before the appearance of damage.

The relative permittivity (ε) and dielectric loss tangent ($\tan \delta$) were measured using a setup consisting of a G4-83 signal generator, an S4-11 spectrum analyzer, and a biconical resonator connected in a transmission configuration. Measurements were carried out at a frequency of 10^{10} Hz and a temperature of 20°C using cylindrical samples ($d=8$ mm, $h=80$ mm).

To evaluate the nonlinear influence of TiO_2 content and sintering temperature on the properties of the experimental ceramics, a full factorial design (3^2) was applied. The investigated factors were TiO_2 content (10–30 weight parts over 100 wt.%) and sintering temperature (1100–1250°C). Experimental data were processed using regression analysis and machine learning methods [11]. Based on the obtained experimental dataset, ML models were developed to analyze the relationships between technological parameters and the properties of the ceramics. Visualization of the results was performed using 2D heatmaps and 3D response surface plots.

Results and discussion

A technological approach enabling the synthesis of dense cordierite ceramics at a reduced temperature of 1350°C was proposed in ref. [12]. The approach is based on replacing part of the components of cordierite ceramics with a relatively low-melting eutectic glass in the pseudo-ternary $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ system. In this case, the cordierite phase is formed during sintering due to the interaction between components of the experimental glass and crystalline fillers.

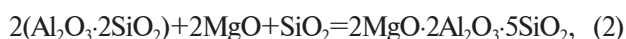
In the present study, the triple eutectic point of the MAS system (1360°C) was selected, at which cordierite, forsterite, and clinoenstatite crystallize. The coefficient of thermal expansion of the MABS glass corresponding to the eutectic composition with the addition of B_2O_3 (10 weight parts over 100 wt.%) is $51.5 \cdot 10^{-7} \text{ }^\circ\text{C}^{-1}$. The crystalline phases clinoenstatite and forsterite are characterized by low dielectric losses ($\varepsilon=6-7$, $\tan \delta=0.0015-0.003$ at 20°C) and relatively high coefficients of thermal expansion, $77.0 \cdot 10^{-7} \text{ }^\circ\text{C}^{-1}$ and $90 \cdot 10^{-7} \text{ }^\circ\text{C}^{-1}$ respectively, and form the basis of radiotechnical ceramics [13]. Considering the more stable physical properties of forsterite compared to clinoenstatite, the possibility of incorporating the $\text{MgO} \cdot \text{SiO}_2$ component of the eutectic glass into the forsterite phase was evaluated.

Titanium dioxide is traditionally introduced into cordierite glass compositions to promote fine

crystallization of the cordierite phase during heat treatment of glass-ceramics. In addition, TiO_2 enables the adjustment of the coefficient of thermal expansion within a desired range, ensuring compatibility between components made of dissimilar materials, and allows maintaining the relative permittivity within specified limits while preserving low dielectric losses [14]. Therefore, possible reactions involving TiO_2 were also considered in the thermodynamic analysis.

The compounds considered as initial components of reactions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system are as follows: MABS glass ($2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$, $2\text{MgO} \cdot \text{SiO}_2$ and $\text{MgO} \cdot \text{SiO}_2$), $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$, SiO_2 , MgO , and TiO_2 .

Thermodynamic calculations of ΔG_T^0 were carried out to assess the feasibility of reactions (1)–(5) during the sintering of ceramics in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system over the temperature range of 873–1473 K.



During thermodynamic calculations, the change in Gibbs free energy of reactions (ΔG_T^0) is determined. Among the possible processes occurring in the system, the most thermodynamically favorable is the one associated with the greatest decrease in ΔG_T^0 .

Thermodynamic constants for minerals that are crystallization products of MABS glass in the glassy state are not available in the literature. Therefore, thermodynamic data for the corresponding crystalline compounds were used. The relatively small differences between the thermodynamic constants of silicates in crystalline and glassy states do not lead to significant changes in the final results.

The values of thermodynamic constants of the initial individual compounds and reaction products at standard temperature are presented in Table 1.

The calculated values of ΔG_T^0 for reactions (1)–(5) in the temperature range of 873–1473 K are presented in Table 2.

Based on the obtained data, in the temperature range of 873–1173 K, the cordierite phase is expected to form first according to reaction (2), as evidenced by the lowest values of ΔG_T^0 ($-493.2 \div -512.9 \text{ kJ} \cdot \text{mol}^{-1}$).

The compounds $\text{MgO}\cdot\text{SiO}_2$ and TiO_2 (rutile), according to the ΔG_T^0 values of reactions (1) and (3)–(5), compete for MgO . Therefore, considering the presence of titanium dioxide in the ceramic composition, it can be expected that, in addition to the crystalline forsterite phase, magnesium titanates, particularly $\text{MgO}\cdot 2\text{TiO}_2$, may also form during sintering.

To verify the results of the thermodynamic calculations, ceramic materials in the $\text{MgO}\text{--}\text{Al}_2\text{O}_3\text{--}\text{SiO}_2\text{--}\text{TiO}_2$ system were synthesized. Titanium dioxide

was introduced in amounts of 10–30 weight parts over 100 wt.%. The chemical compositions of the ceramics are presented in Table 3.

Sintering was carried out in the temperature range of 1100–1250°C, followed by X-ray diffraction analysis of the obtained products (Fig. 1).

It was established that the phase composition of the experimental ceramic CF-5 is represented by α -cordierite ($d\cdot 10^{10}$ =8.30; 4.06; 3.35; 3.11; 3.01; 1.68 m) and forsterite ($d\cdot 10^{10}$ =3.88; 2.89; 2.75; 2.50; 2.44; 1.74 m). With the introduction of titanium

Table 1

Initial thermodynamic constants [15]

Compounds	$-\Delta H_{298.15}^0$, kJ·mol ⁻¹	$-\Delta G_{298.15}^0$, kJ·mol ⁻¹	$S_{298.15}^0$, J·mol ⁻¹ ·K	$C_p = a + b\cdot T + c\cdot T^{-2}$, J·mol ⁻¹ ·K			Temperature range, K
				<i>a</i>	<i>b</i> ·10 ³	<i>c</i> ·10 ⁻⁵	
MgO	601.78	569.53	27.08	42.61	7.28	-6.19	298–2100
MgSiO ₃	1548.92	1462.10	67.86	102.77	19.84	-26.29	298–1800
Mg ₂ SiO ₄	2171.91	2052.93	95.19	149.90	27.38	-35.66	298–1800
α -tridymite	905.98	852.19	43.53	57.10	11.05	0	390–2000
Al ₂ Si ₂ O ₇	3316.15	3102.29	124.24	229.68	36.84	-14.57	298–1173
Mg ₂ Al ₄ Si ₅ O ₁₈	9158.36	8648.19	407.10	602.22	108.0	-161.62	298–1650
TiO ₂ (rutile)	943.87	888.61	50.33	75.20	1.20	-18.20	298–1800
Mg ₂ TiO ₄	2164.01	2047.44	115.10	150.53	35.75	-28.84	298–1800
MgTiO ₃	1571.93	1483.59	74.56	118.42	13.73	-27.33	298–1800
MgTi ₂ O ₅	2507.89	2367.47	135.56	170.29	38.51	-30.77	298–1800

Table 2

Calculated values of ΔG_T^0 for reactions (1)–(5)

Reaction No.	Values of ΔG_T^0 for reactions (kJ·mol ⁻¹) at different temperatures						
	873 K	973 K	1073 K	1173 K	1273 K	1373 K	1473 K
1	-22.5	-22.9	-23.3	-23.8	-24.3	-24.9	-25.5
2	-493.2	-499.8	-506.4	-512.9	–	–	–
3	-26.00	-27.30	-28.69	-30.19	-31.80	-33.54	-35.41
4	-24.29	-24.30	-24.31	-24.39	-24.53	-24.72	-24.97
5	-24.29	-24.91	-25.59	-26.36	-27.24	-28.23	-29.34

Table 3

Chemical compositions of ceramics in the $\text{MgO}\text{--}\text{Al}_2\text{O}_3\text{--}\text{SiO}_2\text{--}\text{TiO}_2$ system (wt.%)

Oxide composition	Composition No.			
	CF-5	CF-5(1T)	CF-5(2T)	CF-5(3T)
SiO ₂	46.65	42.22	38.56	35.48
TiO ₂	0	9.50	17.36	23.95
Al ₂ O ₃	24.11	21.82	19.92	18.34
B ₂ O ₃	4.77	4.32	3.94	3.63
MgO	24.47	22.14	20.22	18.60

dioxide into the ceramic composition, XRD analysis (Fig. 1) clearly reveals, in addition to silicate phases, the presence of titanium-containing phases, particularly anatase ($d \cdot 10^{10} = 3.48; 1.86; 1.63; 1.49$ m) and magnesium dititanate $MgO \cdot 2TiO_2$ ($d \cdot 10^{10} = 4.82; 3.45; 2.73; 1.86$ m), which is consistent with the results of thermodynamic calculations.

Further, the properties of ceramic materials in the $MgO-Al_2O_3-SiO_2-TiO_2$ system were studied in relation to the technological parameters of their processing. To establish functional relationships between processing parameters and material properties, mathematical models were developed using gradient boosting decision tree algorithms implemented in

Python with the *scikit-learn* and *XGBoost* libraries [11]. Such models make it possible not only to generalize experimental data but also to predict ceramic properties within specified parameter ranges, thereby significantly improving the efficiency of composition and processing optimization. The input parameters (features) of the models were TiO_2 content and sintering temperature, while the output parameters (targets) included water absorption, porosity, density, compressive strength, and coefficient of thermal expansion (CTE). The performance of the developed models was evaluated using the coefficient of determination (R^2), which quantifies the proportion of variance in the dependent variable explained by

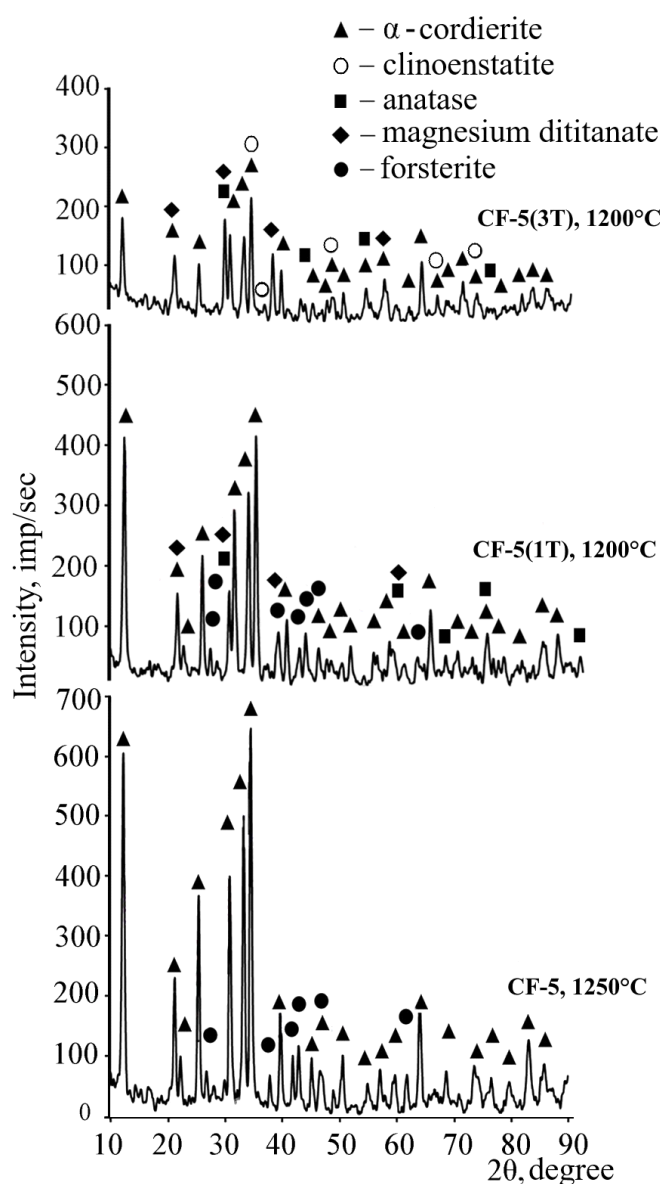


Fig. 1. X-ray diffraction patterns of ceramics in the $MgO-Al_2O_3-SiO_2-TiO_2$ system sintered at different temperatures

the model. For the *XGBoost*-based model, the obtained RI values are presented in Table 4. These results demonstrate good agreement between predicted and experimental values and confirm the adequacy of the ML approach for describing the investigated system.

The feature importance values obtained from the modeling for the properties of the experimental ceramics are presented in Fig. 2.

Feature importance analysis revealed that sintering temperature has a dominant influence (importance values of 0.91–0.99) on the formation of such properties as water absorption, open porosity, apparent density, and compressive strength, whereas the contribution of TiO_2 content is relatively minor. In contrast, an opposite trend is observed for the coefficient of thermal expansion (CTE), for which TiO_2 content is the dominant factor (importance value of 0.99).

For a more illustrative representation of the modeling results, two-dimensional heatmaps and three-dimensional response surface plots were constructed, showing the variation of the physical, mechanical and thermal properties of ceramics in the $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2\text{-TiO}_2$ system as a function of processing parameters (TiO_2 content and sintering temperature) (Figs. 3–7).

It was established that the physical, mechanical and thermal properties of the developed ceramic

materials in the $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2\text{-TiO}_2$ system strongly depend on both TiO_2 content and sintering temperature.

The addition of TiO_2 in the range of 10–30 weight parts over 100 wt.% to the cordierite–forsterite ceramic CF-5 enables the production of a water-impermeable material ($W=0\%$ and $P=0\%$) at a reduced sintering temperature of 1200°C (Figs. 3 and 4). At the same time, the apparent density increases to $2.45\text{--}2.50\text{ g}\cdot\text{cm}^{-3}$ (Fig. 5).

The compressive strength (σ) of ceramic materials in the $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2\text{-TiO}_2$ system increases significantly with increasing sintering temperature from 1100°C to 1200°C , rising from 130–150 MPa to 295–305 MPa.

The coefficient of thermal expansion (CTE) of the experimental ceramics sintered in the temperature range of $1100\text{--}1200^\circ\text{C}$ is primarily governed by the TiO_2 content. With an increase in TiO_2 content from 10 to 30 weight parts over 100 wt.%, the CTE increases from $(31\text{--}32)\cdot 10^{-7}\text{ }^\circ\text{C}^{-1}$ to $(35\text{--}36)\cdot 10^{-7}\text{ }^\circ\text{C}^{-1}$.

For ceramic samples with zero water absorption and zero open porosity, dielectric properties – namely relative permittivity (ϵ) and dielectric loss tangent ($\tan \delta$) – as well as thermal shock resistance were measured (Table 5).

It was established that the developed ceramic materials in the $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2\text{-TiO}_2$ system

Table 4
Coefficients of determination (R^2) of the developed ML models for output parameters

Output parameter (target)	Coefficient of determination (R^2)
Water absorption	0.8263
Open porosity	0.8889
Apparent density	0.6958
Compressive strength	0.9011
Coefficient of thermal expansion (CTE)	0.9351

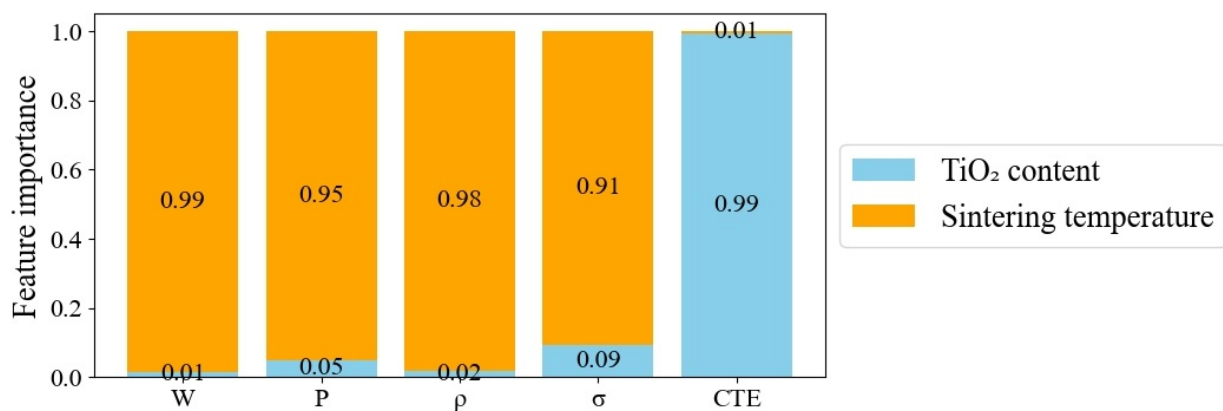


Fig. 2. Relative feature importance of TiO_2 content and sintering temperature for the properties of the experimental ceramics

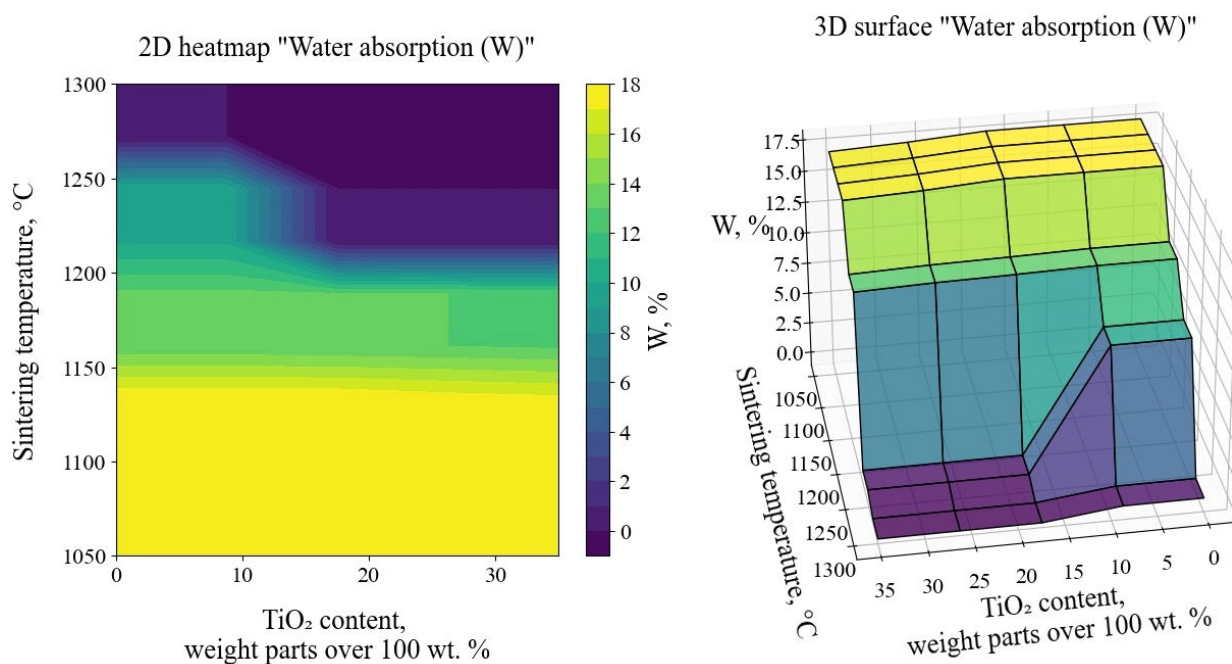


Fig. 3. Two-dimensional heatmap and three-dimensional response surface showing water absorption (%) as a function of TiO₂ content and sintering temperature

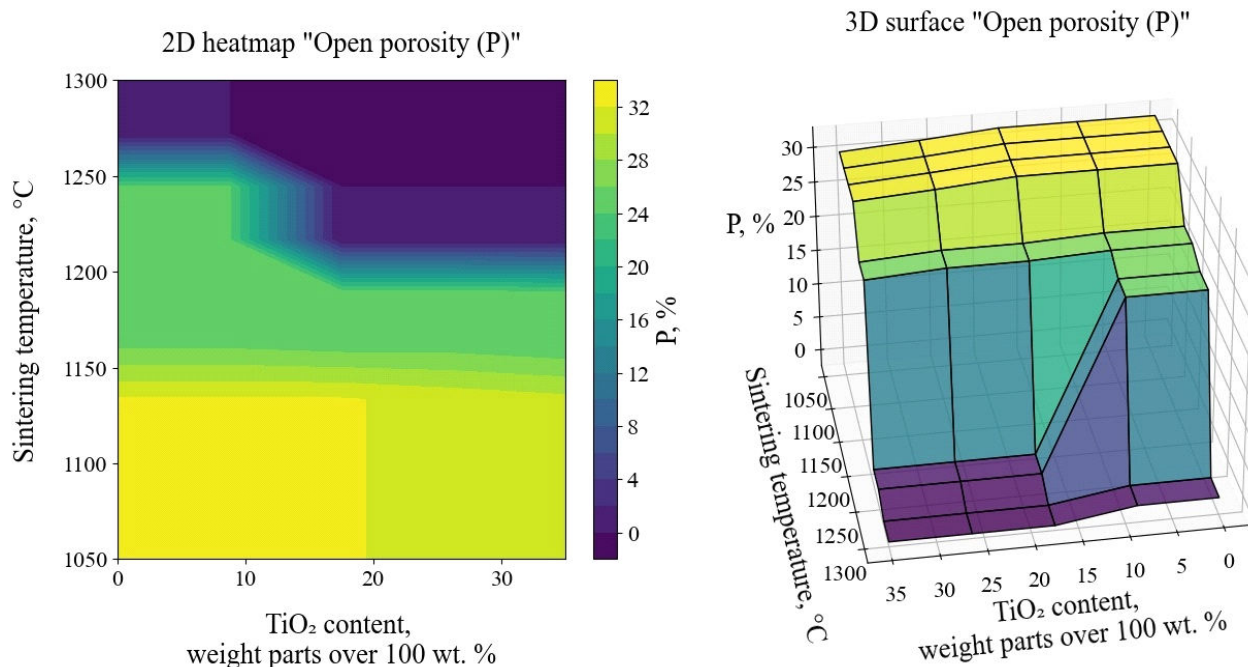


Fig. 4. Two-dimensional heatmap and three-dimensional response surface showing open porosity (%) as a function of TiO₂ content and sintering temperature

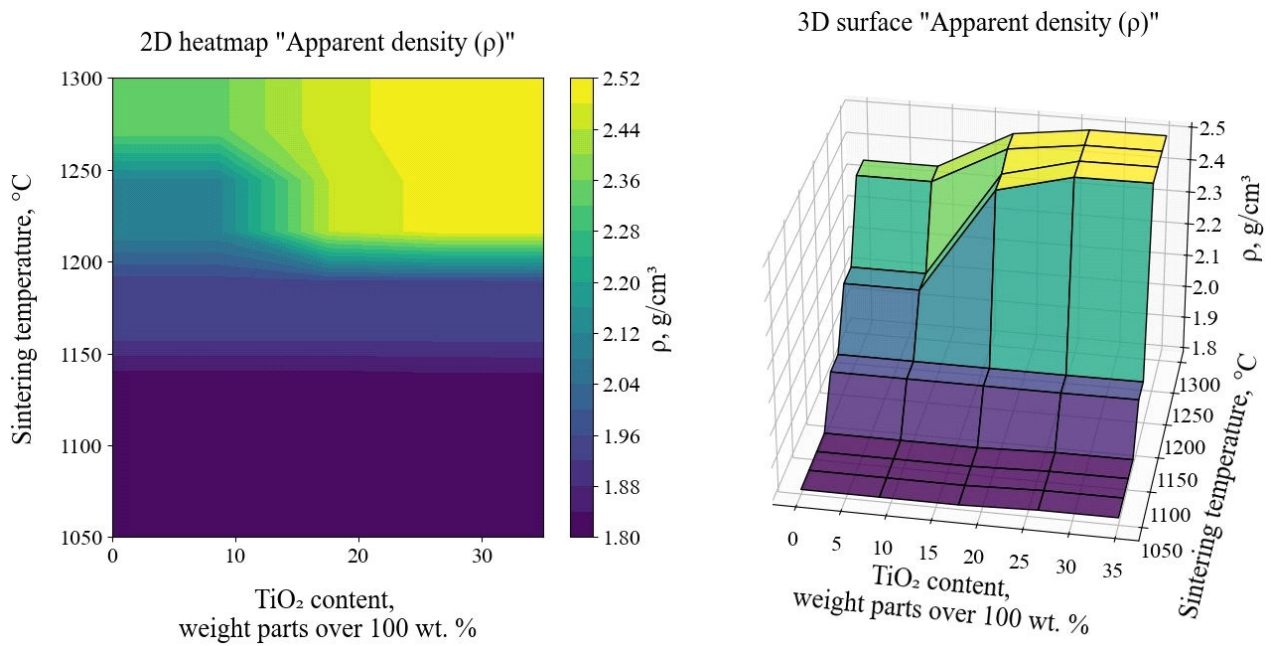


Fig. 5. Two-dimensional heatmap and three-dimensional response surface showing apparent density ($\text{g}\cdot\text{cm}^{-3}$) as a function of TiO₂ content and sintering temperature

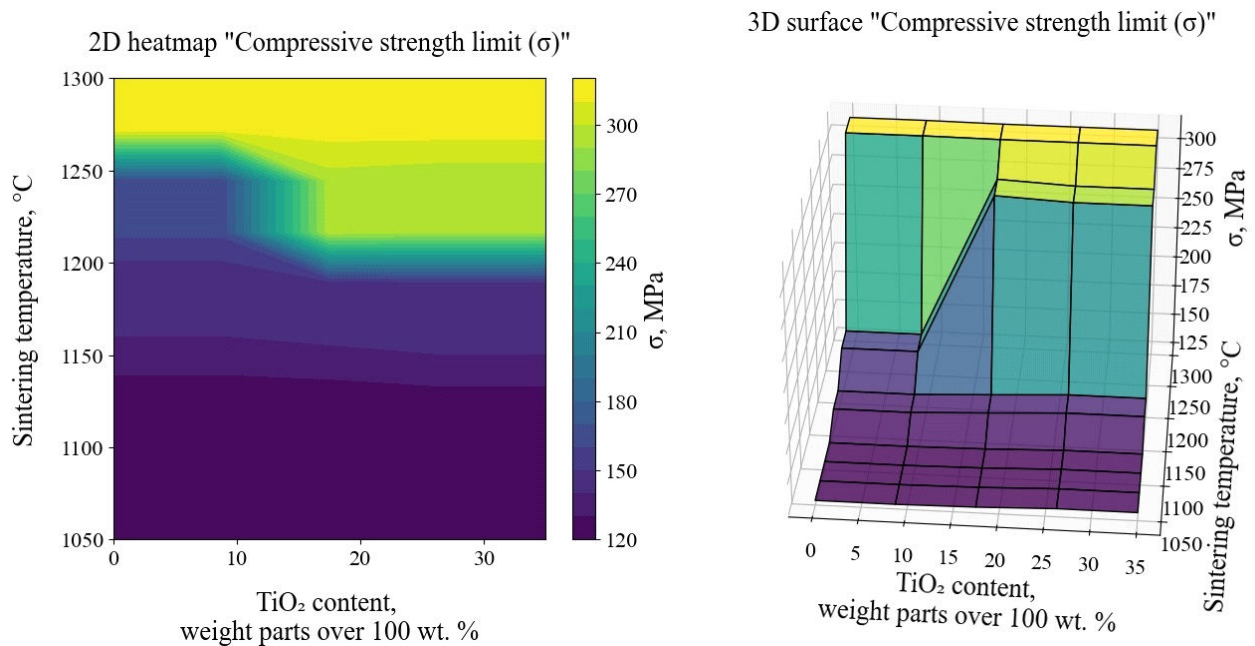


Fig. 6. Two-dimensional heatmap and three-dimensional response surface showing compressive strength (MPa) as a function of TiO₂ content and sintering temperature

exhibit low dielectric losses. The relative permittivity (ϵ) values range from 5.43 to 6.56 and gradually increase with the addition of TiO_2 in the amount of 10–30 weight parts over 100 wt.%. The experimental ceramics are also characterized by low dielectric loss tangent ($\tan \delta$), whose values decrease to 0.0006–0.001 as the TiO_2 content increases up to 30 weight parts over 100 wt.%.

The highest thermal shock resistance (850°C) was observed for the cordierite–forsterite ceramic composition CF-5. The introduction of TiO_2 into the ceramic composition leads to a reduction in thermal shock resistance. At the same time, the thermal shock resistance of ceramics in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system remains at a relatively high level (700–800°C) and meets the requirements for phase shifters used as components of radio-electronic monitoring systems.

Considering the results obtained for the physical, mechanical, thermal, and dielectric properties of the experimental ceramics in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$

system, the most rational combination of properties is achieved at a TiO_2 content of 10–20 weight parts over 100 wt.% and a sintering temperature of 1200°C. Such ceramics exhibit zero water absorption and open porosity, an apparent density of 2.44–2.47 $\text{g}\cdot\text{cm}^{-3}$, and high compressive strength of 295–302 MPa, along with low dielectric losses ($\epsilon=5.8-6.1$; $\tan \delta=0.0008-0.0010$). The coefficient of thermal expansion (CTE) is within $(31-32)\cdot 10^{-7} \text{ }^\circ\text{C}^{-1}$, corresponding to ST-32 glass-ceramics and ensuring high thermal shock resistance (750–800°C).

Conclusions

By introducing part of the components of the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ system in the form of MABS glass, along with the addition of TiO_2 , a densely sintered ceramic material with a combination of functional properties was obtained, enabling its effective application in microwave electromagnetic fields.

A relationship was established between the physical, mechanical, and thermal properties of the

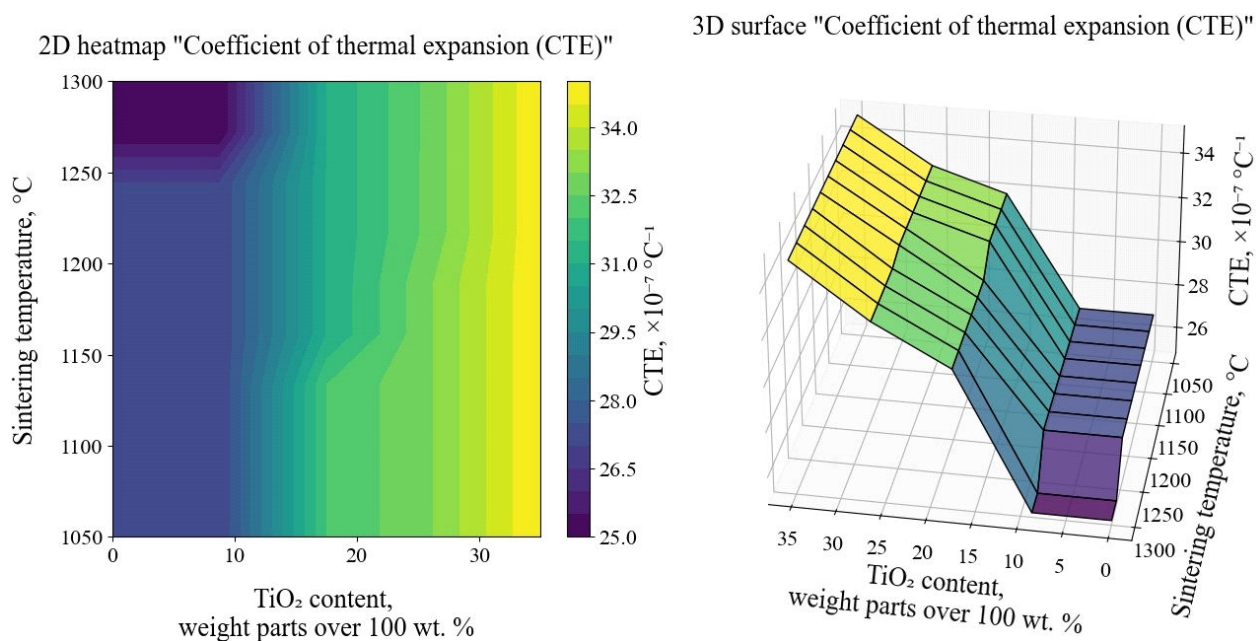


Fig. 7. Two-dimensional heatmap and three-dimensional response surface showing the coefficient of thermal expansion (CTE, $\times 10^{-7} \text{ }^\circ\text{C}^{-1}$) as a function of TiO_2 content and sintering temperature

Table 5

Dielectric properties and thermal shock resistance of experimental ceramics in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system

Composition No.	Sintering temperature, $^\circ\text{C}$	Dielectric properties		Thermal shock resistance, $^\circ\text{C}$
		ϵ	$\tan \delta$	
CF-5	1250	5.43	0.0013	850
CF-5 (1T)	1200	5.77	0.0010	800
CF-5 (2T)	1200	6.10	0.0008	750
CF-5 (3T)	1200	6.56	0.0006	700

experimental ceramics and the technological parameters of their processing (sintering temperature and TiO_2 content). Corresponding mathematical models were developed using gradient boosting decision tree algorithms implemented in Python with the *scikit-learn* and *XGBoost* libraries. The application of machine learning methods made it possible to quantitatively assess the relative contribution of controlled factors and to enhance the prediction of material properties within the investigated parameter range.

The most rational ceramic compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system, in terms of achieving a favorable combination of physical, mechanical, thermal, and dielectric properties, were identified. These compositions enable sintering at the relatively low temperature of 1200°C . The resulting combination of properties makes the developed ceramics suitable for use as microwave materials in components of radio-electronic monitoring systems, particularly phase shifters.

Conflict of interest

O.V. Zaichuk is an Editorial Board Member of the journal but was not involved in the peer-review process or the decision-making regarding this manuscript. All other authors declare no conflict of interest.

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РАДІОПРОЗОРИ КЕРАМІЧНІ МАТЕРІАЛИ НА ОСНОВІ СИСТЕМИ $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$: ФІЗИЧНІ, МЕХАНІЧНІ, ТЕРМІЧНІ І ДІЕЛЕКТРИЧНІ ВЛАСТИВОСТІ

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Виробництво фазообертачів фазованих антенних решіток, як правило, здійснюється з кордієритових ситалів марок СТ-32 і СТ-38 чи аналогічної за складом склокераміки. Такі матеріали повинні володіти водонепроникністю, малими діелектричними втратами у НВЧ-діапазоні, заданими значеннями температурного коефіцієнта лінійного розширення і високими термо-механічними властивостями. Робота присвячена розробці складів низькотемпературної кераміки в системі $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ для виготовлення фазообертачів з регульованими фізичними, механічними, термічними і діелектричними властивостями. Для оцінки перебігу можливих реакцій в системі $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ за участю компонентів скла евтектичного складу в системі $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ були проведені термодинамічні розрахунки. Розглянута точка потрійної евтектики в системі $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$, в якій кристалізуються наступні фази: кордієрит $2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$, форстерит $2\text{MgO}\cdot\text{SiO}_2$ і кліноенстатит $\text{MgO}\cdot\text{SiO}_2$. При проведенні термодинамічного аналізу визначали зміни енергії Гіббса для можливих реакцій утворення кордієритової і форстеритової фази за участю компонентів скла евтектичного складу, а також реакцій за участю TiO_2 . Встановлено взаємозв'язок фізичних, механічних і термічних властивостей дослідної кераміки з технологічними параметрами її виготовлення (температурою випалу, вмістом TiO_2). Використання методів машинного навчання дозволило кількісно оцінити відносний внесок керованих факторів і розширити можливості прогнозування властивостей розробленої кераміки в межах інтервалу параметрів, які досліджувались. Визначені найбільш раціональні з точки зору досягнення комплексу високих показників властивостей складу кераміки в системі $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$, які дозволяють проводити її випал при низькій температурі 1200°C.

Ключові слова: радіопрозора кераміка, фазообертач, кордієрит, евтектичне скло, титан(IV) оксид, термодинамічний аналіз, випал, фізико-механічні властивості.

RADIO-TRANSPARENT CERAMICS IN THE $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ SYSTEM: PHYSICAL, MECHANICAL, THERMAL AND DIELECTRIC PROPERTIES

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The production of phase shifters for phased-array antennas is typically based on cordierite glass-ceramics, such as grades ST-32 and ST-38, or on materials of similar composition. Such materials are required to exhibit water resistance, low dielectric losses in the microwave frequency range, controlled values of the coefficient of thermal expansion, and high thermomechanical performance. This study focuses on the development of low-temperature ceramic compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system for the fabrication of phase shifters with tailored physical, mechanical, thermal, and dielectric properties. Thermodynamic calculations were performed to evaluate possible reactions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system involving components of eutectic glass compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ system. The triple eutectic point of this system was considered, at which the following phases crystallized: cordierite ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$), forsterite ($2\text{MgO}\cdot\text{SiO}_2$), and clinoenstatite ($\text{MgO}\cdot\text{SiO}_2$). The thermodynamic analysis included the determination of Gibbs free energy changes for possible reactions leading to the formation of cordierite and forsterite phases, as well as reactions involving TiO_2 . A relationship between the physical, mechanical, and thermal properties of the developed ceramics and the technological parameters of their processing (sintering temperature and TiO_2 content) was established. Machine learning methods made it possible to quantitatively assess the relative contributions of the controlled factors and to improve the prediction of the properties of the developed ceramics within the investigated parameter range. The most rational compositions in the $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{TiO}_2$ system were identified, providing a favorable combination of properties and enabling sintering at 1200°C.

Keywords: radio-transparent ceramics; phase shifters; cordierite; eutectic glass; titanium dioxide; thermodynamic analysis; sintering; physical and mechanical properties.

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