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INORGANIC SYNTHESIS AND INDUSTRIAL INORGANIC CHEMISTRY

Solid-Phase Synthesis and Dielectric Properties of Materials Based on $LaAlO_3$–$CaTiO_3$ System

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Abstract—Effect of synthesis conditions on the formation and dielectric characteristics of materials based on the $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ phase was studied. It is shown that the new approach suggested in the study to obtaining ceramic materials based on the $LaAlO_3$–$CaTiO_3$ system via introduction of an excess amount of Al_2O_3 over the stoichiometry of $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ results in that a finely grained material is formed with relative permeability $\varepsilon = 45$, temperature coefficient of the relative permittivity $\tau_\varepsilon = -66 \times 10^{-6}$ K^{-1}, dielectric losses $\tan \delta = 2 \times 10^{-4}$, and quality factor $Q = 4700$ at $f = 9$ GHz. The functional characteristics of the resulting material are comparable with the parameters of the materials synthesized by chemical coprecipitation and indicate that its use in elements of microwave electronics is promising.

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The synthesis of dielectric ceramic materials based on the $LaAlO_3$–$CaTiO_3$ system for electronic elements has been the subject of a number of studies [1–8]. It has been shown that it is necessary to choose a technology that provides the full formation of solid solutions of the main crystalline phase under the industrially possible variations of the grinding and calcination conditions, choice of starting reagents, etc. Such an approach has been found by using the “wet chemistry” method in manufacture of these compositions, which made it possible to lower the temperature at which the xLaAlO$_3$–$(1 - x)$CaTiO$_3$ solid solutions are formed to 1150°C and to reach stable and reproducible dielectric characteristics, as considered in [4, 7–9]. However, this method requires an expensive technology and it is rather difficult to control with this method the structure of the composite being formed and, accordingly, also the electrical properties of the resulting materials, as noted in [10–12]. Therefore, an important practical task is to find approaches that would satisfy the condition of obtaining stable dielectric characteristics in production of materials by solid-phase synthesis.

An analysis of published data obtained in studies of the solid-phase processes in which materials are formed on the basis of xLaAlO$_3$–$(1 - x)$CaTiO$_3$ compositions (x = 0.1, 0.3, 0.5, 0.7, 0.9) demonstrated that, as the content of lanthanum aluminate is raised to more than 50 mol %, a processing temperature as high as 1550°C is necessary for obtaining a material of prescribed composition [1, 2]. According to a number of publications [3, 5, 13], materials with the component ratio 0.7CaTiO$_3$: 0.3LaAlO$_3$ possess the most promising dielectric characteristics. However, it was noted in [1–6] that obtaining materials with the component ratio xLaAlO$_3$: $(1 - x)$CaTiO$_3$ (x = 0.1, 0.3) in the temperature range 1300–1500°C presents difficulties associated with the incomplete occurrence of the solid-phase reaction in which a solid solution of required composition is formed and by-products affecting the functional characteristics are produced. According to [2], the dielectric characteristics of the material of the xLaAlO$_3$–$(1 - x)$CaTiO$_3$ (x = 0.3) system, sintered at 1550°C, are $Q \times f = 36071$ GHz and $\varepsilon_r = 36.6$. It was noted in [1] that a solid-phase synthesis of the material based on the xLaAlO$_3$–$(1 - x)$CaTiO$_3$ (x = 0.325) system at

Table 1. Modes of solid-phase synthesis of the material based on the $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ phase

Method of synthesis	Solid-phase synthesis conditions		
	starting components	thermal-treatment mode	
		synthesis temperature, °C/exposure, h	sintering temperature, °C/exposure, h
1	$CaCO_3$, La_2O_3, Al_2O_3, TiO_2	1100–1400/3	1530/3
2	$CaTiO_3$ and $LaAlO_3$	1440, 1530/3	1530/3
3	$CaTiO_3$ and ($LaAlO_3$ + 2 wt % Al_2O_3)	1580/3	1530/3

temperatures of 1380–1450°C and varied chemical state yields various secondary compounds are formed and the $Q \times f$ values change to 35 000–38 000 GHz.

The goal of our study was to find how synthesis conditions affect the formation and dielectric properties of material based on the $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ phase.

EXPERIMENTAL

We used three variants of the solid-phase synthesis of materials based on phases with the perovskite structure, $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$. Specific features of the solid-phase synthesis of phases with the perovskite structure were described in detail in [1–3]. Let us specify the formation conditions of the materials under consideration, which includes choosing the starting components and the thermal treatment modes (Table 1).

In the first variety of the solid-phase synthesis (method 1), samples were produced from simple oxides. As the starting reagents served $CaCO_3$ (99.5 wt %, average grain size (D) = 2 μm), La_2O_3 (>99.8 wt %, D = 1.0 μm), Al_2O_3 (>99 wt %, D = 1.0 μm), and TiO_2 (99.3 wt %, D = 0.5 μm).

In the second method, the material was synthesized from a mixture of $CaTiO_3$ and $LaAlO_3$. When $LaAlO_3$ was produced from simple lanthanum and aluminum oxides, the synthesis temperature was varied (1440 and 1530°C).

The third method for synthesis of the material based on the $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ phase differs from method no. 2 in that an excess amount of Al_2O_3 (2 wt %) is introduced over the stoichiometry of $LaAlO_3$ in its synthesis from simple lanthanum and aluminum oxides.

Analytical methods. The qualitative and quantitative phase compositions of the samples obtained were monitored by X-ray diffraction (XRD) analysis with a Shimadzu XRD-7000 diffractometer (CuK_α radiation). The peaks were identified and the phase ratio was determined from the XRD patterns of the samples under study with a PDWin 4.0 software package.

The microstructure and the phase composition of sample nos. 1–3 (Table 2) were determined by scanning electron microscopy (SEM) and energy-dispersive X-ray fluorescence elemental microanalysis on a Quanta 200 scanning electron microscope equipped with an EDAX microprobe attachment. The error in determining the content of the elements by the given method varies with the atomic number of an element, being about 0.3 wt % on average. The microprobe attachment was calibrated against the spectra of La, Ca, Al, Ti, and O, measured from the reference disk shipped by Oxford Instruments. The average grain size and the grain size distribution were determined by using Thixomet Lite image-analysis software.

The pycnometric density of the samples was determined with an Ultrapycnometer 1000 helium pycnometer. The X-ray density was calculated on the basis of the data obtained for unit cell parameters of the phases synthesized in the study. The unit cell parameters of the $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ phases produced by different variants of solid state synthesis were calculated for the orthorhombic crystallographic system accordingly the data presented in [3, 5, 14].

The capacitance (C) and the dielectric losses (tan δ) were determined with an MTsE-17 device at a frequency of 1 MHz on samples with deposited electrodes. The relative permittivity was found by calculation on the basis of the experimental data obtained by measuring the capacitance of test samples at a frequency of 1 MHz. The relative permittivity was determined in the microwave range by the method of a waveguide-dielectric resonator. The natural resonance frequency and the quality factor of disk dielectric resonators were determined with a VDR-DRD device by the radial-line method.

Table 2. Results of X-ray fluorescence microanalysis for regions shown in Figs. 1b–3b

Material	Region being analyzed	Content of components according to the analysis, mol %				Phase
		$AlO_{1.5}$	CaO	TiO_2	$LaO_{1.5}$	
1	SQ	18.93	32.65	32.65	15.77	$34.60LaAlO_3–65.40CaTiO_3$
	P1	16.67	33.12	33.77	16.42	$33.40LaAlO_3–66.60 CaTiO_3$
	P2	18.72	32.37	33.16	15.75	$34.47LaAlO_3–65.53CaTiO_3$
	P3	17.47	32.66	33.88	15.98	$33.48LaAlO_3–66.52CaTiO_3$
	P4	18.27	32.19	32.93	16.60	$34.87LaAlO_3–65.13CaTiO_3$
	P5	18.35	32.50	33.62	15.53	$33.88LaAlO_3–66.12CaTiO_3$
			12.12	54.54	33.33	Phase close to $La_4Ca_2Ti_5O_{18}$
2	SQ	19.81	31.54	32.11	16.54	$36.35LaAlO_3–63.65CaTiO_3$
	P1	18.33	32.03	32.73	16.91	$35.24LaAlO_3–64.75CaTiO_3$
	P2	17.08	31.94	33.62	17.36	$34.44LaAlO_3–65.56CaTiO_3$
	P3	19.19	31.54	32.69	16.58	$35.77LaAlO_3–64.23CaTiO_3$
	P4	20.42	31.91	31.55	16.09	$36.51LaAlO_3–63.49CaTiO_3$
	P5	17.01	32.77	32.99	17.23	$34.24LaAlO_3–65.76CaTiO_3$
		16.15			83.85	Phase close to $LaAl_{11}O_{18}$
3	SQ	15.33	33.59	33.97	17.10	$32.43LaAlO_3–67.57CaTiO_3$
	P1	16.38	33.13	33.32	17.00	$33.38LaAlO_3–66.62CaTiO_3$
	P2	17.80	32.10	32.59	17.51	$35.31LaAlO_3–64.66CaTiO_3$
	P3	15.71	33.90	34.17	16.22	$31.93LaAlO_3–68.07CaTiO_3$
	P4	12.29	36.31	29.69	21.70	$34.02LaAlO_3–65.98CaTiO_3$
	P5	16.74	33.26	33.47	16.53	$33.27LaAlO_3–66.73CaTiO_3$

RESULTS AND DISCUSSION

The phase of the $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ composition was produced from the oxides $CaCO_3$, La_2O_3, Al_2O_3, and TiO_2.

Analysis of the XRD data (Fig. 1) indicates that, after the starting mixture of the oxides is calcined at 110°C for 3 h, the $CaTi_4O_9$ and La_2O_3 phases and a phase close in composition to $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ are recorded. The formation of a solid solution close in composition to $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ under these conditions is presumably due to the activation of processes corresponding to the transformation of a surface (2D nonautonomous) phase to the liquid state [10, 15], which causes an active interaction between the individual compounds $CaTiO_3$ and $LaAlO_3$, formed at lower treatment temperatures. The appearance of the $CaTi_4O_9$ phase is presumably due to the incomplete reaction of $CaTiO_3$ formation via the interaction of the reactive CaO produced as a result of the decarbonization of $CaCO_3$ in the temperature range 620–930°C [16] with TiO_2.

As follows from the data in Fig. 1, raising the treatment temperature to 1200°C results in the formation of a by-product, $La_4Ca_2Ti_5O_{18}$ phase. It is noteworthy that the following oxides Al_2O_3, $CaTi_4O_9$, La_2O_3 are formed together with the target product (Fig. 1a). The formation of the synthesis by-product $La_4Ca_2Ti_5O_{18}$ can

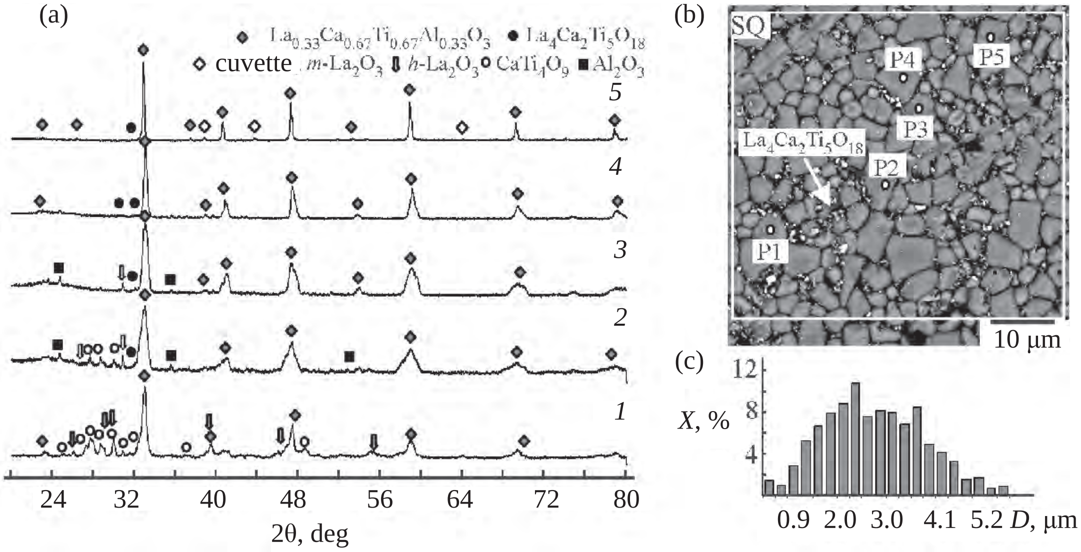

Fig. 1. (a) (*1–4*) XRD patterns of samples synthesized from a mixture of the simple oxides CaO, TiO_2, La_2O_3, Al_2O_3 at temperatures in the range 1100–1400°C and (*5*) XRD pattern; (b) SEM data; and (c) particle size distribution for the $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ material synthesized from a mixture of the simple oxides CaO, TiO_2, La_2O_3, Al_2O_3. (2θ) Bragg angle, (*X*) fraction of grains, and (*D*) grain size; the same for Figs. 2 and 3.

be attributed to the reaction between $CaTi_4O_9$, $CaTiO_3$, and La_2O_3:

$$CaTi_4O_9 + CaTiO_3 + 2La_2O_3 = La_4Ca_2Ti_5O_{18}.$$

As follows from the data in Fig. 1a, raising the treatment temperature to 1300 and 1400°C, leads to a decrease in the intensity of the reflections attributed to the oxides $CaTi_4O_9$ and La_2O_3, as well as to the formation of the $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ phase with large degree of crystallinity.

According to XRD data (Fig. 1), the target phase $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ with large degree of crystallinity is recorded after the synthesis from a mixture of the simple oxides $CaCO_3$, TiO_2, La_2O_3, and Al_2O_3 at 1530°C for 3 h. A quantitative XRD analysis of sample no. 1 demonstrated the presence of 96 mol % $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ phase and 4 mol % $La_4Ca_2Ti_5O_{18}$ phase (Fig. 1a). According to SEM and elemental analysis data (Fig. 1b, Table 2), the $La_{0.35}Ca_{0.65}Ti_{0.65}Al_{0.35}O_3$ phase (SQ, Table 2) close in the solid-solution composition to that set in synthesis, $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$, after the synthesis from a mixture of the simple oxides $CaCO_3$, TiO_2, La_2O_3, Al_2O_3 at 1530°C for 3 h. Also noteworthy is the presence of the compound $La_4Ca_2Ti_5O_{18}$ with a perovskite-like structure, recorded by the methods of XRD and X-ray fluorescence microanalysis, which is localized between grains of the main phase (Figs. 1a and 1b).

It is possible to preclude formation of the by-product of synthesis, $La_4Ca_2Ti_5O_{18}$, by changing the reaction pathway, i.e., by affecting the chemical prehistory. This can be done, e.g., by using other starting components, changing the dispersity of the reagents and the homogeneity of the reaction formulation, introducing heterophase additives and a small excess of a reagent, and using other methods that change the formation order and type of intermediate products of the solid-phase interaction.

Synthesis of $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ from preliminarily obtained compounds $CaTiO_3$ and $LaAlO_3$. Because it was shown that the by-product $La_4Ca_2Ti_5O_{18}$ is formed when $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ is produced from the simple oxides CaO, TiO_2, La_2O_3, Al_2O_3, a new approach was suggested. This approach consists in that the target product is synthesized from the preliminarily obtained individual compounds $CaTiO_3$ (Fig. 2a) and $LaAlO_3$. The procedure for obtaining $LaAlO_3$ was described in Experimental.

Figure 2a shows an XRD pattern of a $LaAlO_3$ sample produced by thermal treatment of a starting mixture of lanthanum and aluminum oxides at 1440°C for 3 h. It should be noted that the target product, $LaAlO_3$, is formed under the specified conditions and the phases of lanthanum hydroxide and lanthanum oxide that crystallizes in the cubic modification c-La_2O_3 are recorded. The appearance of the c-La_2O_3 and $La(OH)_3$ phases, determined by

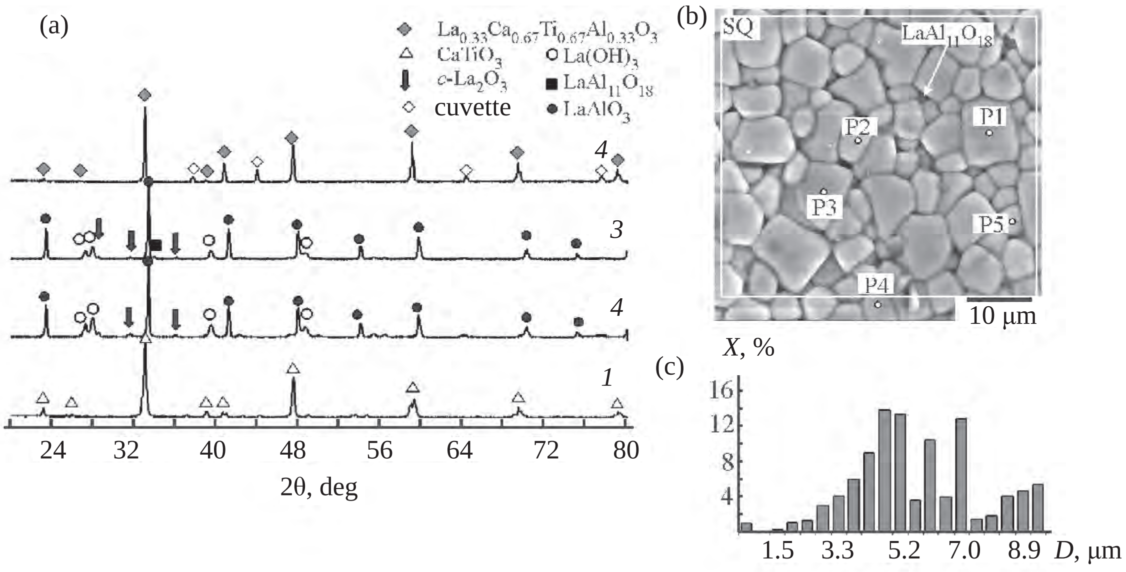

Fig. 2. (a) XRD patterns of (*1*) $CaTiO_3$ sample and $LaAlO_3$ sample after the thermal treatment in air at (*2*) 1440 and (*3*) 1530°C at the stoichiometry of the $LaAlO_3$ compound, and (*4*) XRD pattern; (b) SEM data; and (c) particle size distribution for the $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ material synthesized from $CaTiO_3$ and $LaAlO_3$.

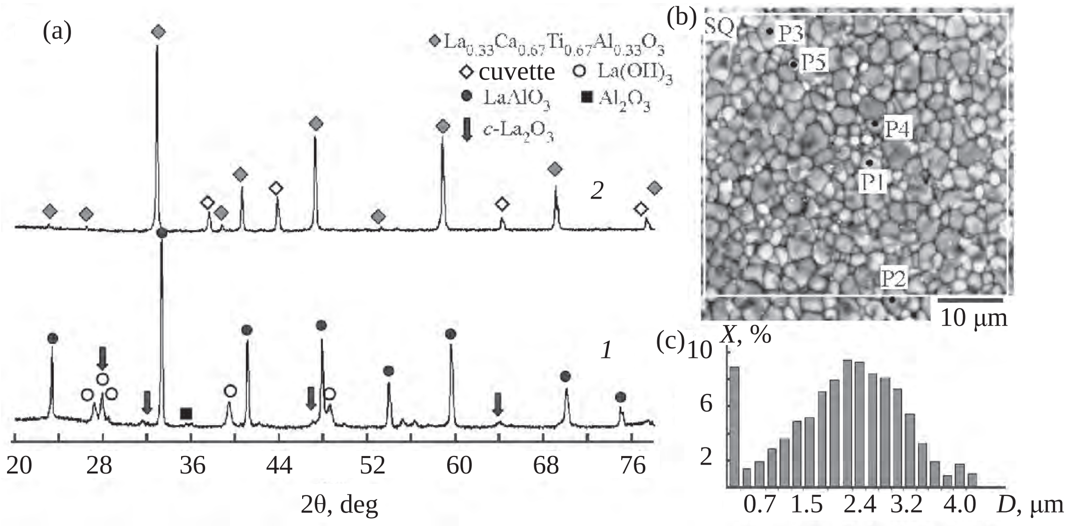

Fig. 3. (a) (*1*) XRD pattern of the oxide $LaAlO_3$ obtained with addition of a 2 wt % excess of Al_2O_3 after the synthesis by the ceramic technology at 1580°C and (*2*) XRD pattern; (b) SEM data; and (c) particle size distribution for the $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ material synthesized from $CaTiO_3$ and $LaAlO_3$ obtained with addition of a 2 wt % excess of Al_2O_3.

the conditions in which the formulations are prepared (grinding and mixing with water in air and the pronounced tendency of La_2O_3 to undergo hydration and carbonization [17]), is presumably due to the incomplete occurrence of the reaction in which $LaAlO_3$ is formed. The fact that a heterogeneous sample is obtained when the phase is synthesized on the basis of $LaAlO_3$ may be due to the hindered mass transfer of the starting oxides across the blocking layer of the reaction product, which requires, in particular, that higher temperatures should be used. Raising the $LaAlO_3$ synthesis temperature to 1530°C results both in a decrease in the intensity of the reflections related to lanthanum oxide and in the appearance in trace amounts of the oxide $LaAl_{11}O_{18}$ (b-Al_2O_3), in agreement with the data of [17, 18] (Fig. 2a).

The microstructure of the material after its synthesis at 1530°C in the course of 3 h from the compounds $CaTiO_3$ and $LaAlO_3$ that is produced by solid-phase synthesis at

Table 3. Parameters of the crystal structure and X-ray, pycnometric, and relative density of materials 1–3

Material	*a*, Å ± 0.001	*b*, Å ± 0.006	*c*, Å ± 0.008	*V*, Å^3	$r_{\text{X-ray}}$	r_{pycn}	r_{rel}, %
					g cm^{-3}		
1	5.428	5.429	7.698	226.88	4.78	4.72	98.74
2	5.415	5.422	7.661	224.94	4.84	4.63	95.66
3	5.432	5.448	7.684	227.40	4.72	4.61	97.66

Table 4. Electrical characteristics of materials 1–3

Material	*T*, °C	Measurements at *f* = 1 MHz		Microwave measurements			
		ε	τ_ε × 10^6, K^{-1}	ε	tan δ	*Q*	*Q* × *f*, GHz
1	1530	46.6	–16	44.7	1.8 × 10^{-4}	4073	37000
2		47.5	–37	44.3	2.3 × 10^{-4}	3220	30000
3		48.5	–66	45.3	2.2 × 10^{-4}	4530	42000

1530°C is shown as an example in Fig. 2b. According to the data in Table 2, the sample under consideration contains a mixture of solid solutions with compositions close to that of the target product to be synthesized. Also noteworthy is the insignificant deviation of the ratio between lanthanum and aluminum oxides from 1 : 1 (Table 2), which presumably points to the incomplete formation of the $LaAlO_3$ phase, as regarded in [18]. It is also necessary to note the presence of the $LaAl_{11}O_{18}$ phase localized between the grains of the main phase.

Synthesis of $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ from $CaTiO_3$ and $LaAlO_3$ that is obtained with addition of a 2 wt % excess of Al_2O_3. The results of the XRD analysis of a sample produced by thermal treatment of a mixture of simple lanthanum and aluminum oxides with addition of a 2 wt % excess of Al_2O_3 over the stoichiometry of the target compound $LaAlO_3$ at 1580°C for 3 h are presented in Fig. 3a. According to the data in Fig. 3a, the $LaAlO_3$ sample synthesized at 1580°C contains the main phase $LaAlO_3$ and small amounts of the starting reagents La_2O_3 and Al_2O_3.

According to the results of an XRD analysis (Fig. 3a), the solid-phase synthesis of the target product from $CaTiO_3$ and $LaAlO_3$ obtained with addition of a 2 wt % excess of Al_2O_3 at 1530°C in the course of 3 h yields the $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ phase with large degree of crystallinity.

The microstructure of the ceramic after its synthesis at 1530°C for 3 h is shown in Fig. 3b. According to the results of an elemental analysis (Table 2), the material under consideration corresponds, within the error of the method used, to the component ratio set in the synthesis. It should be noted for the material that a finely grained ceramic is formed, with the grain size not exceeding 4 μm. Presumably, this result should be attributed to the pronounced hindrance to grain growth, which occurs, e.g., in the presence of insoluble impurities, particles of the second phase, etc. The introduction of an excess amount of Al_2O_3, which is a heterophase additive in synthesis of $LaAlO_3$, results in the localization of the latter at grain boundaries and thereby makes slower the grain growth process.

Characterization and dielectric properties of materials. The nature of the grain size distribution, found from SEM data for materials 1–3, is shown in Fig. 1c–3c. The average grain sizes in the grain size distributions for compositions 1–3 are 2.5, 5.5, and 2.0 μm, respectively (Figs. 1c–3c). For compositions 1 and 3, a more uniform particle size distribution is observed, with predominance of fractions with particle size smaller than 0.3 μm for material 3.

The results obtained in determining unit cell parameters and the X-ray, pycnometric, and relative densities for materials 1–3 are presented in Table 3. Analysis of the data in Table 3 shows that the pycnometric densities for materials 1–3 are 4.61–4.72 g cm^{-3}, in good agreement with the X-ray density calculated from XRD data. It is noteworthy that the largest relative density is observed for materials 1 and 3 (Table 3).

According to the data in Table 4, compositions 1–3 have about the same values of the relative permittivity ($\varepsilon \approx 45$ in the microwave range and $\varepsilon \approx 46$ at $f = 1$ MHz) and dielectric losses ($\tan \delta \approx 2 \times 10^{-4}$), whereas the temperature coefficients of the permittivity and the quality factors are different.

The value of $Q \times f$ for composition 2 has a smaller value (30 000 GHz) as compared with these characteristics for materials 1 and 3. This presumably results from the presence of both a by-product ($LaAl_{11}O_{18}$) and a mixture of solid solutions with composition close to that of the target product formed in the synthesis, which is presumably indicative of the incomplete occurrence of the solid-phase reaction.

The value of $Q \times f$ for sample 1 is 37 000 GHz. Presumably, the presence of the $La_4Ca_2Ti_5O_{18}$ phase in the sample as the by-product of the solid-phase synthesis affects the quality factor. According to [19], $Q \times f$ for $La_4Ca_2Ti_5O_{18}$ does not exceed 29 000 GHz.

The maximum value of Qf (42 000 GHz) is observed for material 3. The temperature coefficient of the relative permittivity for material 3 is close to the values for the material obtained by sintering of complex oxides synthesized by the chemical coprecipitation method [8]. Thus, the best dielectric properties are observed for material 3, which can be accounted for by the formation of a homogeneous solid solution of composition $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ and by the synthesis of a fine-grained ceramic material with high relative density. The data obtained indicate that the technology described in the present communication is promising for application in elements of microwave electronics.

CONCLUSIONS

Thus, the formation processes and the dielectric properties of the $La_{0.33}Ca_{0.67}Al_{0.33}Ti_{0.67}O_3$ composition produced by solid-phase synthesis from simple oxides La_2O_3, Al_2O_3, TiO_2, CaO and from a mixture of $LaAlO_3$ and $CaTiO_3$ were considered. It was shown that, in all the cases under study, the $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ phase is produced via formation of individual compounds $LaAlO_3$ and $CaTiO_3$ through heterovalent substitution without changes in the total number of atoms in the unit cell of $CaTiO_3$.

It was demonstrated that the relative permittivity and the dielectric losses of $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ ceramic materials produced by solid-phase synthesis from $CaCO_3$, La_2O_3, Al_2O_3, and TiO_2, $LaAlO_3$ and $CaTiO_3$ and with addition of an excess amount of Al_2O_3 over the stoichiometry of the target product to be synthesized have close values of 45 and 2×10^{-4}, respectively. It was shown that the values of $Q \times f$ at a frequency of 9 GHz for materials that are based on the $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ phase and are obtained by three variants of the solid-phase synthesis are 30 000, 37 000, and 42 000. It was found that the introduction of an excess amount of Al_2O_3 over the stoichiometry for $La_{0.33}Ca_{0.67}Ti_{0.67}Al_{0.33}O_3$ results in that a finely grained material is formed with a temperature coefficient of the relative permittivity of -66×10^{-6} K^{-1}. The functional characteristics of the material are comparable with the parameters of materials synthesized by the chemical coprecipitation method, which indicates that it is promising for application in elements of microwave electronics.

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