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Influence of ultrasonic mechanoactivation on phase formation in dielectric ceramic material based on $BaAl_2Si_2O_8$

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1. Introduction

Rapid development of electronics requires using materials of high quality. Research on the development of inorganic dielectrics with desired properties has become increasingly important. The important factor in process of obtaining material is the possibility of formation of a required crystal structure, in particular, using ultrasonic mechanoactivation [1].

$BaAl_2Si_2O_8$ shows complex polymorphism and crystallizes in monoclinic, orthorhombic and hexagonal structures [2–5]. The physical properties of celsian depend on its crystal structure. For example, a compound with the monoclinic structure is thermodynamically stable in the temperature range of 20–1590°C [5], whereas the hexagonal phase of celsian is stable up to 1760°C [1].

Low thermal shock resistance that related to the structural transformation of the hexagonal α-modification into the hexagonal β-modification in the 280–320°C temperature range appears to be a drawback of hexacelsian [6].

Monoclinic form does not undergo polymorphic transformations and is characterized by high dielectric and mechanical properties.

The aim of this work is to study the influence of ultrasonic mechanoactivation on crystal structure of $BaAl_2Si_2O_8$.

2. Experimental procedures

$BaAl_2Si_2O$ was obtained by using the oxides of $BaCO_3$, Al_2O_3, SiO_2 as the initial components in the ratio of 1:1:2. The synthesis was carried out in alundum crucibles in air by solid-state reaction at temperatures of 1300–1450°C, the time of synthesis was 2 h. The synthesized powders were wet-ground in ethanol. The coupling agent (e.g., PVA glue) was added to some parts of synthesized powder after which that then pressed at 100 MPa into pellets. The sintering was carried out at temperatures from 1440°C to 1520°C for 2 h in air atmosphere.

Another part of the powder was ultrasonically treated at normal atmospheric pressure for 0.5–1.5 h using ultrasonic generator UZG 1-1 of 1 kW power and magnetostrictive transducer PMS 1-1. Then obtained material was pressed at 100 MPa into pellets and sintered at temperatures from 1250°C to 1350°C for 2 h in air atmosphere. The phase composition of the obtained material after processes of synthesis, ultrasonic mechanoactivation and sintering was determined by X-ray diffraction metod of monochromatized CuK_α radiation in the angle range of 20–65°. Dielectric measurements of the samples were made at 100 kHz with an E7-8 bridge in the temperature range of 20–350°C. Open porosity was studied using the optical microscope Olympus GX 41. Processing of the experimental data was performed using the software Autoscan 2500 Studio.

3. Results and discussion

According to X-ray diffraction studies ceramics of hexagonal $BaAl_2Si_2O$ is obtained after synthesis (the presence of the monoclinic modification is recorded only against a background (Fig. 1)).

The analysis of X-ray diffraction patterns shows that after sintering the crystal structure is similar to the initial one for the samples obtained from the synthesized material without mechanoactivation (Fig. 1).

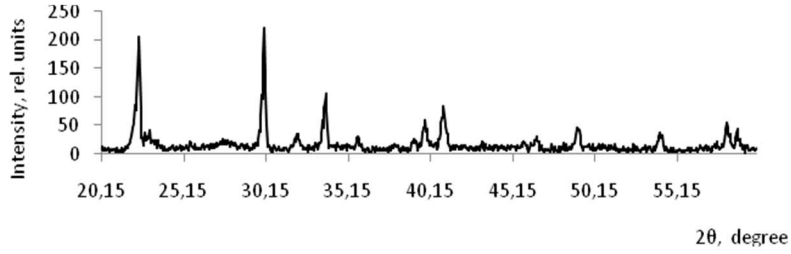

Fig. 1. X-ray diffraction pattern of ceramic samples of $BaAl_2Si_2O$ after synthesis

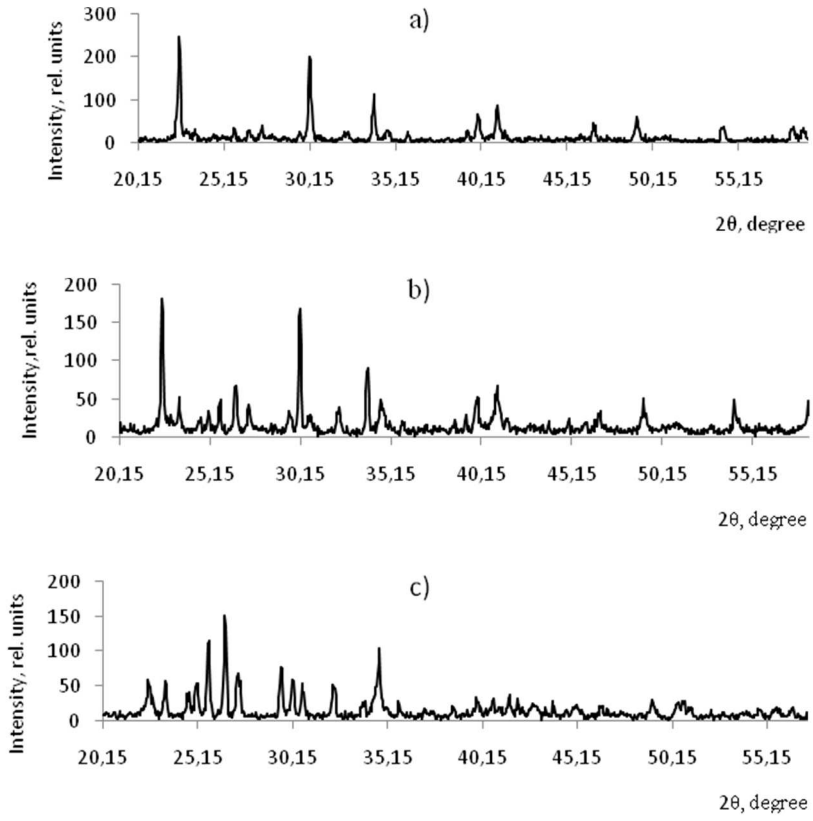

Fig. 2. X-ray diffraction pattern of ceramic sample of hexagonal $BaAl_2Si_2O$ after ultrasonic treatment and subsequent heat treatment: *a*) UVs treatment for 0.5 h; *b*) UVs treatment for 1 h; *c*) UVs treatment for 1.5 h

It is found that the crystal structure of hexagonal modification was formed at synthesis temperatures of 1300–1450°C. For single-phase ceramic samples of hexagonal $BaAl_2Si_2O$ the temperatures of synthesis and sintering must be 1450°C and 1500°C, respectively.

The study of the samples obtained after mechanoactivation (UVs grinding) and subsequent sintering shows that the influence of ultrasonic vibrations (UVs) for 0.5 h (Fig. 2a) leads to a monoclinic modification. The influence of UVs leads to the increase of crystal structure of the monoclinic modification (Fig. 2b).

Further increase in ultrasonic exposure time for 1.5 h results in the formation of monoclinic crystal structure (Fig. 2c).

The results of studies of electrophysical properties of the samples with different types of crystal structure are shown in Table 1.

Table 1

Properties	Hexagonal modification	Monoclinic modification	Phase mixture
Relative dielectric constant, *e*	6.5±0.2	6.3±0.2	6.4±0.2
Dielectric loss tangent, tg δ	no more 0.0005	no more 0.0005	no more 0.0005
Temperature coefficient of permittivity, TK e 10^{-6}, 1/ºC	106	109	106
Porosity, %	5.61	6.45	6.55

The temperature dependence of the dielectric constant of the samples with different modifications of crystal structure shows that in the temperature range from 20°C to 290°C the dielectric behavior does not depend on the modification of crystal structure.

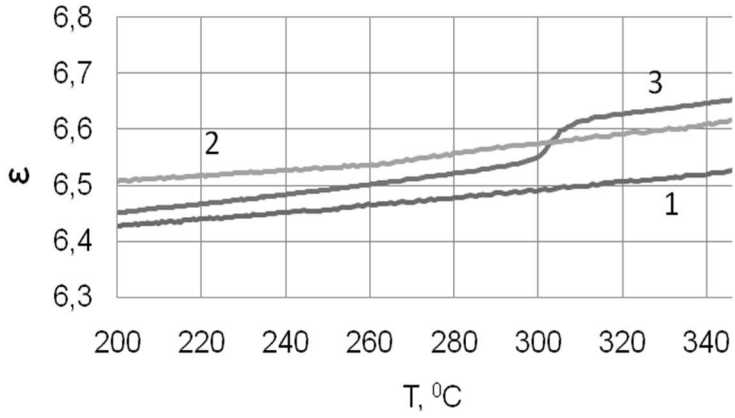

Fig. 3. Temperature dependencies of permittivity of the samples with different modifications of crystal structure: 1 – monoclinic, 2 – mixture of phases, 3 – hexagonal

Structural transformation of hexagonal α-modification into β-modification for ceramic samples of $BaAl_2Si_2O$ occurs in the temperature range of 280–320°C that is coherent with the results of DTA analysis [8], where the transition temperature corresponds to 312°C. Monoclinic form has not polymorphic transformations. The phase transition for the two-phase samples is lack (Fig. 3).

Resume

The studies have shown that hexagonal $BaAl_2Si_2O$ is synthesized at temperatures of 1300–1450°C. Subsequent ultrasonic treatment of the synthesized $BaAl_2Si_2O$ stimulates polymorphic transformations, i.e. by selecting the ultrasonic treatment regime it is possible to obtain a material with the given modification of crystalline structure. Thus, an increase in processing time of UVs up to 1.5 h leads to the formation of the monoclinic-phase structure.

It has been determined that the values of electrical parameters of the sample $BaAl_2Si_2O$ do not depend on the modification of crystalline structure. This ceramic material has low porosity, high Q-factor and good dielectric parameters that allows to use it for ceramic resonators, and other microwave devices.

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Abstract

Mechanoactivation effect has been studied on crystal structure of $BaAl_2Si_2O_8$ known as polymorphous compound. Ultrasonic treatment of the synthesized material powder has been used as mechanoactivation method.

Ultrasonic grinding during 0.5–1.5 h was determined to result in polymorphous transformation.

Dielectric properties of ceramics $BaAl_2Si_2O_8$ have been investigated for hexagonal, monoclinic modifications of crystal structure as well for that based on phase mixture.

It is shown that sintering of ceramic material based on monoclinic crystal structure modification of $BaAl_2Si_2O_8$ takes place in temperature diapason of 1300–1350ºC.

Ceramic materials based on $BaAl_2Si_2O_8$ compounds have been shown to exhibit low porosity, high Q-factor and dielectric parameters allowing use them for resonators, and other microwave devices.

Streszczenie

Zbadano właściwości magnetyczne anionowo zubożonego kobaltytu $La_{1-x}Ba_xCoO_{3-\delta}$ ($x \geq 0.5$)

Pole magnetyczne powoduje bardzo duże zmiany w oporności wraz z dużą histerezą magnetyczną. Ustalono że wywierany naprężenia zmienia stan antyferromagnetyczny na ferromagnetyczny. Przyjęto się, że podstawowy stan magnetyczny silnie zależy od objętości komórki jednostkowej oraz że przejścia są związane z tym że jony kobaltu odwracają kierunek obrotu. Sugeruje się, że stan antyferromagnetyczny odpowiada wysokiemu stanowi spinu jonów kobaltu podczas kiedy ferromagnetyczny jest wynikiem pośredniego stanu spinowego jonów kobaltu.