UDC 534.29-7/-8; 621.762.4; 666.655

DEVELOPMENT OF LEAD-FREE PIEZOCERAMICS WITH APPLICATION OF ULTRASONIC VIBRATIONS

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Abstract

The development of lead-free ferroelectric materials is an important task due to the restrictions on the use of lead in industry enacted in a number of European countries, the United States, and Japan (Directive 2011/65/EU of the European Parliament). In the Republic of Belarus, a technical regulation TR EAEU 037/2016 has been in effect since 2018, established by the EAEU member states. This technical regulation sets down requirements for the restriction of the use of hazardous substances in electrotechnical and radio-electronic products.

The bismuth sodium titanate solid solution system Na0.5Bi0.5TiO3 (NBT) is considered one of the promising materials for practical application, despite its characteristics being inferior to those of the classical PZT-system. The application of ultrasonic vibrations during the synthesis of PZT-ceramics suggests that ultrasound at different stages of the production of NBT-based piezoceramics could also enhance its physical and mechanical properties.

This work presents the results on the influence of ultrasonic vibrations during mechanoactivation and pressing on the properties of lead-free piezoceramics of the NBT type. It has been shown that applying ultrasonic vibrations during mechanoactivation and pressing leads to the grinding of the initial powder, allowing the obtaining of denser homogeneous ceramics, preventing the formation of microcracks during sintering, reducing the sintering temperature by ~60° C, increasing the Curie temperature by 50–60° C, and increasing the dielectric constant. Ceramics produced with ultrasound is characterized by high electromechanical anisotropy (d33/d31), which is important for practical applications when it is necessary to excite certain vibration modes. The obtained results will serve as a basis for further development of the scientific field of ultrasonic mechanoactivation and pressing of piezoceramic powders.

Keywords: piezoceramics, ultrasound, ultrasonic mechanoactivation, sodium bismuth titanate, ultrasonic pressing.

ПОЛУЧЕНИЕ БЕССВИНЦОВОЙ ПЬЕЗОКЕРАМИКИ С ПРИМЕНЕНИЕМ УЛЬТРАЗВУКОВЫХ КОЛЕБАНИЙ

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Реферат

Разработка сегнетоэлектрических материалов, не содержащих свинец, является актуальной задачей, что обусловлено ограничениями по использованию свинца в промышленности, принятыми в ряде Европейских стран, США и Японии (директива Европарламента 2011/65/EU). В Республике Беларусь с 2018 г. действует технический регламент ТР ЕАЭС 037/2016, разработанный странами, входящими в Таможенный союз, который запрещает использование токсичных элементов в радиоэлектронной промышленности.

Система твердых растворов титаната-натрия-висмута Na0.5Bi0.5TiO3 (NBT) считается одной из перспективных в плане практического применения несмотря на то, что по своим характеристикам она уступает классической PZT-системе. Результаты применения ультразвуковых колебаний в процессе синтеза PZT-керамики позволяют предположить, что применение ультразвука на различных этапах производства пьезокерамики на основе NBT способно также улучшить ее физико-механические свойства.

В данной работе показано влияние ультразвуковых колебаний на этапах механоактивации и прессования на свойства бессвинцовой пьезокерамики типа NBT. Установлено, что ультразвуковая механоактивация и прессование с наложением ультразвуковых колебаний приводит к измельчению исходного порошка, что позволяет получать более плотную однородную керамику, предотвратить образование микротрещин при спекании, снизить температуру спекания на ~60 °C и повысить температуру Кюри на 50–60 °C, увеличить диэлектрическую проницаемость. Для керамики, полученной с применением ультразвука, характерна высокая электромеханическая анизотропия (d33/d31), что важно для практического применения, когда необходимо возбуждать определенные моды колебаний. Полученные результаты послужат основой для дальнейшего развития научного направления ультразвуковой механоактивации и прессования пьезокерамических порошков.

Ключевые слова: пьезокерамика, ультразвук, ультразвуковая механоактивация, титанат натрия висмута, прессование с ультразвуком.

Introduction

A study [1] on the compaction of structural zirconia ceramics using ultrasonic compaction demonstrated the possibility of obtaining products with high strength characteristics, which are established at the stage of producing press blanks from ultrafine powders. This significantly reduces the influence of powder characteristics such as bulk density, flowability, formability, and compactibility without the use of lubricants and binding additives. Studies on the ultrasonic pressing of ferroelectric and piezoelectric ceramic powders [2–6] also demonstrated that the finished products have higher density and improved physical and mechanical properties compared to those obtained using traditional technology.

The traditional method of compacting powdered parts in a metal mold [7] involves loading the powder into a die and compacting it by applying high static pressure. A disadvantage of this method is the highly uneven distribution of mechanical stresses throughout the volume, which can lead to subsequent failure of the pressed or sintered part. To increase static pressure on powders of hard and brittle materials causes high elastic stresses at the contact points between particles and brittle fracture in these areas when the pressure is released that makes it impossible to form large parts with complex geometric shapes. The use of plasticizers does not completely solve the problem due to the formation of a significant number of pores when the powder burns out during sintering.

One effective method for modifying the properties of ceramic materials is the use of powerful ultrasonic vibrations at different stages of their production: ultrasonic mechanoactivation of powder, pressing, and synthesis. Ultrasonic mechanoactivation is based on the phenomenon of acoustic cavitation – formation and collapse of cavities in a liquid medium being subjected to ultrasonic waves. In this process, particles and their powder aggregates are crushed as a result of repeated exposure to pulsed loads caused by the collapse of cavitation bubbles. Furthermore, grinding is possible due to the collision of powder particles during their random movement under the influence of ultrasonic vibrations. Due to mechanoactivation the sintering temperature of the processed powder decreases, the mechanical properties improve (reduced porosity, increased strength, improved plastic properties), and reactivity increases [8–11].

In ultrasonic powder pressing, high-power ultrasound is typically applied to the powder using a punch or a die via the entire compaction cycle or by applying a certain pressure from the beginning of the process. The principle of ultrasonic pressing is that through the periodic application of ultrasonic vibrations with an amplitude comparable to the size of particles or their agglomerates, the overall stress in the deformed powder material is uniformly redistributed and relaxes within the compact due to the uniform distribution of powder particles. When pressing with ultrasonic vibrations, the pressure distribution within the compact becomes more uniform due to both reduced wall friction and reduced interparticle friction, resulting in denser particle packing and an increased density of the pressed blank. Furthermore, the use of ultrasound during pressing results in the healing of small pores and a reduction in the number and size of large pores [12].

Thus, a major advantage of using ultrasound in the process of pressing is to produce compacts without a mechanical stress gradient, as otherwise, subsequent operations will result in product failure due to the formation of delamination cracks. Reducing pressing pressure is generally not considered the primary advantage of using ultrasonic vibrations. Firstly, the application of ultrasonic vibrations requires additional energy consumption. Secondly, higher compact density can be achieved, and is much easier, by increasing pressure. However, pressure can be increased up to a certain optimal value. For example, for barium titanate, this value is 30÷70 MPa [12]. Exceeding the optimal pressure causes cracking. This is because, as pressing pressure increases, the elastic force causing cracking increase proportionally to the pressure, while the strength of the compact increases only slightly. As a result, when pressing pressure reaches a certain value, the destructive elastic strength exceeds the strength of the compacts, and cracks appear in the pressed product.

Based on the described physical laws of the pressing process, it is proposed [13] to initially press to the optimal pressure value without applying ultrasonic vibrations. Then, to relieve uneven mechanical stress in the sample, apply ultrasonic vibrations to the compacted mass in a static state under the optimal fixed pressure.

Applying ultrasonic vibrations to the pressed powder for less than one second is ineffective, since the energy of ultrasonic vibrations in this case is insufficient to relieve the uneven distribution of mechanical stress that arises during the pressing process. The application of ultrasound for 2–5 s leads to complete equalization of stresses throughout the entire volume of the pressed product. Increasing the ultrasonic exposure time beyond 5 s does not improve the quality of the pressed product and reduces the service life of the press mold [13].

Employing rotational movements in the press tooling (matrix and punch) can enhance the quality of press blanks. The analysis of the dependence of powder density on pressing pressure for barium titanate showed that the application of ultrasonic vibrations increases the density of samples obtained at the same pressure. This difference is particularly significant at low pressure.

Studies of the compaction process of $BaTiO_3$ powder using holographic interferometry methods revealed that at the initial stage of pressing without ultrasonic vibrations, compaction occurs in a limited area near the upper punch and then extends to the middle and lower parts of the compact. As the compaction density increases, particle motion becomes more orderly, and slippage decreases. When pressing with ultrasound, the powder undergoes the same compaction stages as without ultrasound vibrations and, when it reaches a state of density, behaves as a monolithic body. However, when pressing with ultrasound at lower pressures, the powder reaches a state where the particles begin to move in a regulated manner, reducing contact friction forces between the powder particles and between the powder and the die walls, providing the desired density with lower static stress [14].

The aim of this work is to study the influence of ultrasonic vibrations on the physical and mechanical properties NBT – NBT-BT based piezoceramics.

Experimental procedure

Ultrasonic mechanoactivation of powder can be achieved using an immersible sonotrode or by exciting vibrations in a container (cup) where the powder is loaded (Figure 3). Powder processing can be performed dry, i.e., without pouring it into a liquid medium, or by placing the powder in a liquid in which ultrasonic vibrations are excited. In the first case, the physics of the process are similar to powder processing in planetary mills. In the second case, mechanoactivation and crushing of powder agglomerates occur through ultrasonic dispersion in a liquid medium due to cavitation and ultrasonic flows. Moreover, the efficiency of ultrasonic processing is significantly increased if the process is carried out under excess hydrostatic pressure (Figure 2). Excess hydrostatic pressure in a process medium was created using a compressor.

Two acoustic schemes were used for pressing the synthesized powder with the application of ultrasonic vibrations: the half-wave and quarter-wave reflectors (Figure 4). The first scheme (Figure 4a) is convenient because the amplitude of displacements at the end of the half-wave reflector can be seen to estimate the amplitude of vibrations in the powder deformation (compacting) zone. A laboratory setup was designed and built for these schemes using a UZDN-2T generator with a power of 0.4 kW, a frequency of ~22 kHz, and a manual hydraulic press with a force of 35 kPa. This setup was used to test the compaction tests.

The charge was prepared from Na_2CO_3, TiO_2 and Bi_2O_3 powders. Mixing was performed in a jasper mortar using alcohol. The powder was then pressed into briquettes of 20 mm in diameter and 10 mm in height for subsequent synthesis. The synthesis temperature was 800–850 °C, and the synthesis time was 5 hours: a 3-hour temperature rise and a 2-hour hold. To minimize deviations from the calculated stoichiometric composition due to evaporation of volatile oxides, the synthesis was carried out in a closed crucible with a backfill of powder of the same composition. Samples of composition (1-x) NBT – xBT, where x = 0,1; 0,12, were obtained using a similar technology.

The X-ray diffraction pattern of the obtained compound (Figure 1) showed a typical perovskite structure [15], as a confirmation of a successful synthesis.

The synthesized briquettes were ground using a mortar. The stoichiometric mixtures of the initial NBT and NBT – BT powders obtained after synthesis were then subjected to ultrasonic mechanoactivation using a 2 kW ultrasonic generator operating at a frequency of ~22 kHz in distilled water for 30 minutes (Figures 2, 3).

For ultrasonic pressing of the synthesized powder we used a 0,4-kW generator with a frequency of ~22 kHz and a manual hydraulic press (Figures 4, 5). The pressing force was 35 kPa.

The obtained press blanks were sintered in a closed crucible at 1150 °C for 5 hours. Then the end surfaces of the samples were ground to ensure flatness and parallelism for subsequent application of conductive coatings and dielectric measurements. Electrodes were applied by firing silver paste using a standard method at 800 °C. The dielectric measurements were carried out using an E7-20 immittance meter at a frequency of 1 kHz.

The study of granulometric composition of powders was analyzed using the Malvern Mastersizer 2000 laser analyzer (Malvern Instruments Ltd., Malvern, UK) in the range of 0,02–2000 μm according to the standard method.

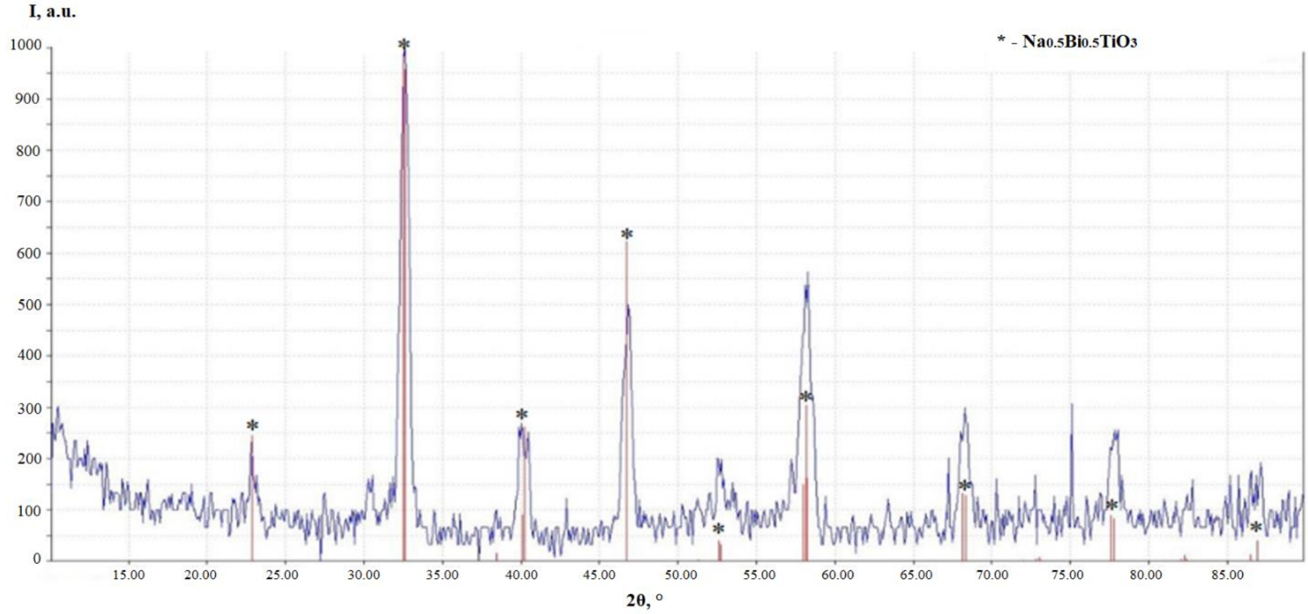
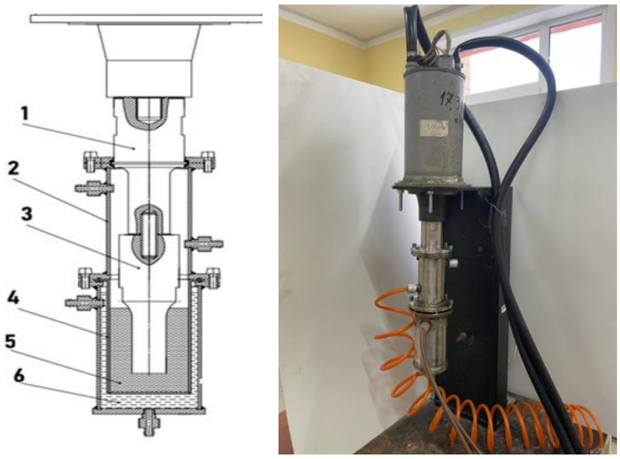


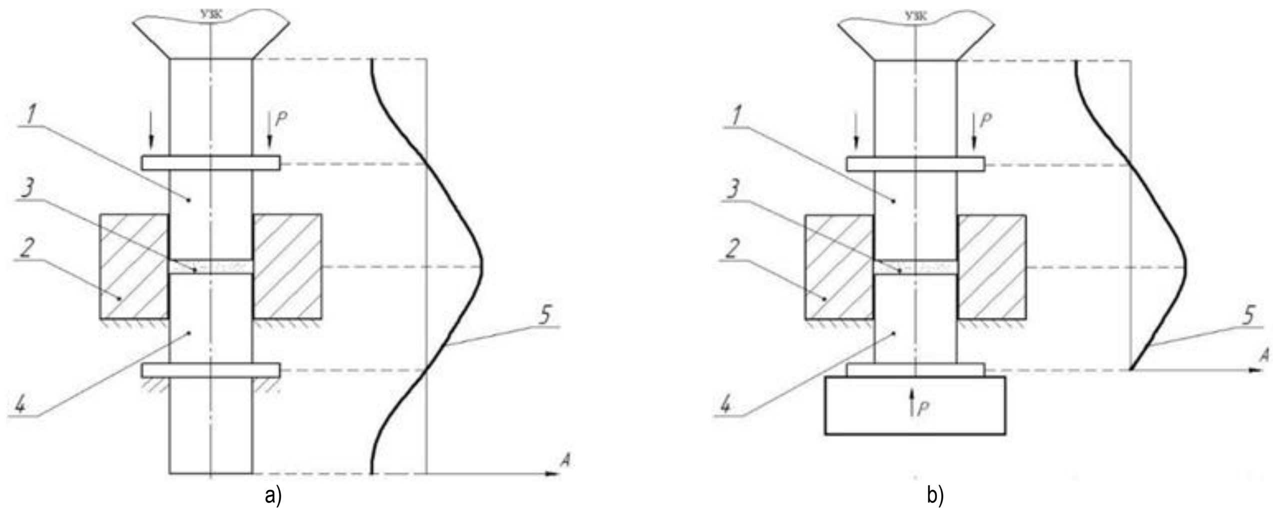
Figure 1 – X-ray diffraction pattern of the studied NBT (*– peaks of the standard substance from the database)



1 – booster; 2 – reactor chamber body; 3 – waveguide; 4 – glass;
5 – processed medium; 6 – cooling liquid

Figure 2 – Scheme and acoustic unit for ultrasonic mechanoactivation of powders:

Figure 3 – Laboratory setup for ultrasonic mechanoactivation



1 – half-wave waveguide-punch; 2 – matrix; 3 – powder; 4 – reflector; 5 – diagram of mechanical displacements

Figure 4 – Schematic diagram of an acoustic unit with a half-wave (a) and quarter-wave-reflector (b) for ultrasonic pressing of powder

https://doi.org/10.36773/1818-1112-2024-135-3-104-110

Figure 5 – Experimental setup for ultrasonic powder pressing

Results and discussion

The results of particle size analysis (Figures 6 and 7) showed that the ultrasonically treated powder reduced particle agglomeration and improved particle size. Thus, while the average particle size of the original powder was 67,5 μm, the average particle size after ultrasonic treatment was 5,7 μm.

Figure 8 shows pictures of powder before and after ultrasonic mechanoactivation.

At the next stage we studied the microstructure of sintered press blanks obtained both without the application of ultrasonic vibrations and with ultrasonic vibrations.

The ultrasonic-assisted pressing process leads to a reduction in the number of large pores and a more uniform structure of the press blank (Figure 9). Usually, the grain-size, grain orientation and porosity, can strongly affect the domain structure and polarization switching behavior [16–18].

Measurement of the permittivity showed that the application of ultrasonic vibrations at different stages of the synthesis of NBT-based piezoceramics leads to an increase in the permittivity (Figure 10). Curves are strongly frequency-dependent, which is characteristic of a relaxor-like behavior [19], potentially arising from disordered polar regions [20].

The permittivity increased for samples of each composition obtained using ultrasonic vibrations. The maximum increase in permittivity was observed for the composition with x = 0,12. The increase in the dielectric loss tangent values for the samples subjected to ultrasonic vibrations was observed compared to those for the samples obtained using a standard technology. However, in the temperature range from 370 to 400 °C, the tangent values for the composition with x = 0,12 obtained using ultrasonic vibrations were lower than those for the samples of the same composition obtained using a standard technology (Figure 11).

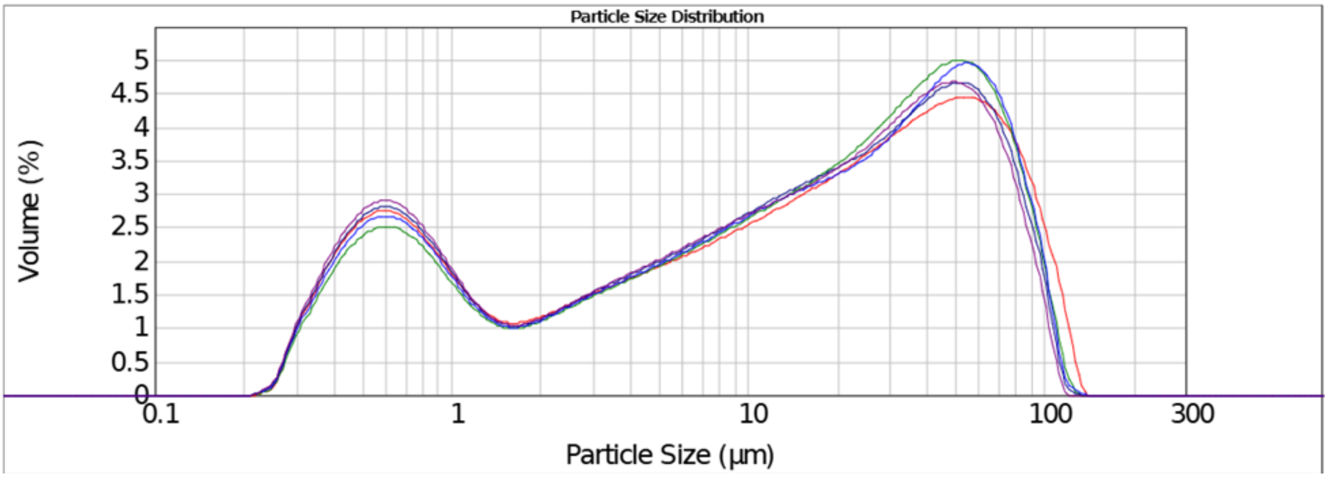


Figure 6 – Particle size distribution of the initial powder

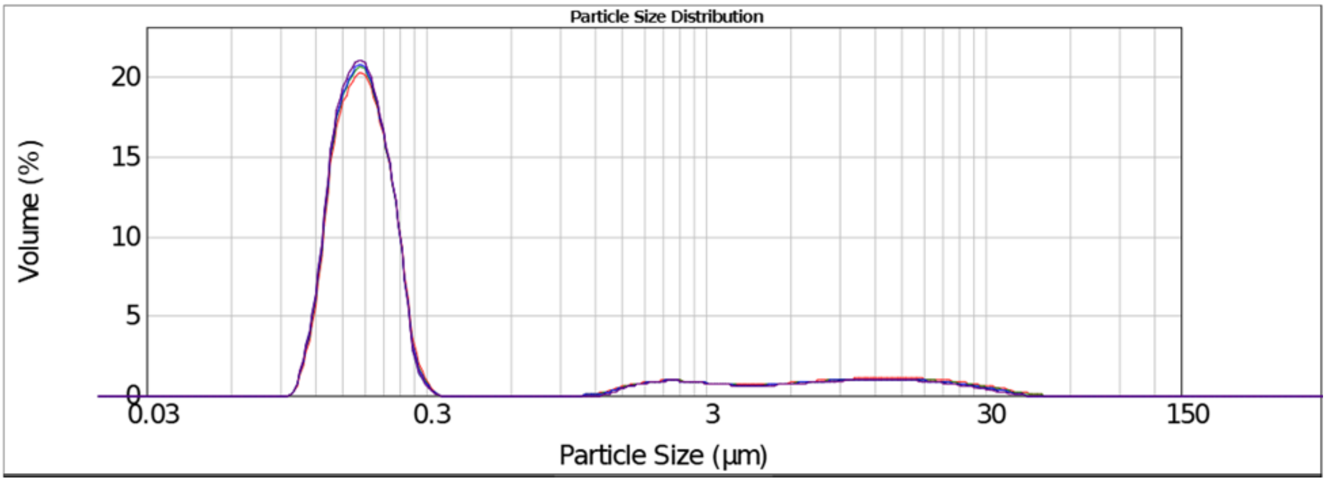
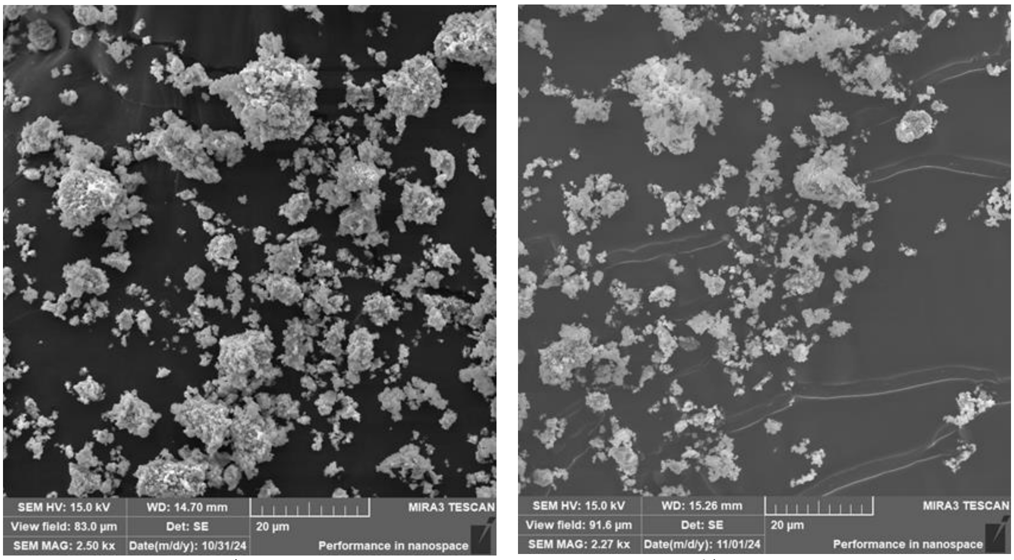
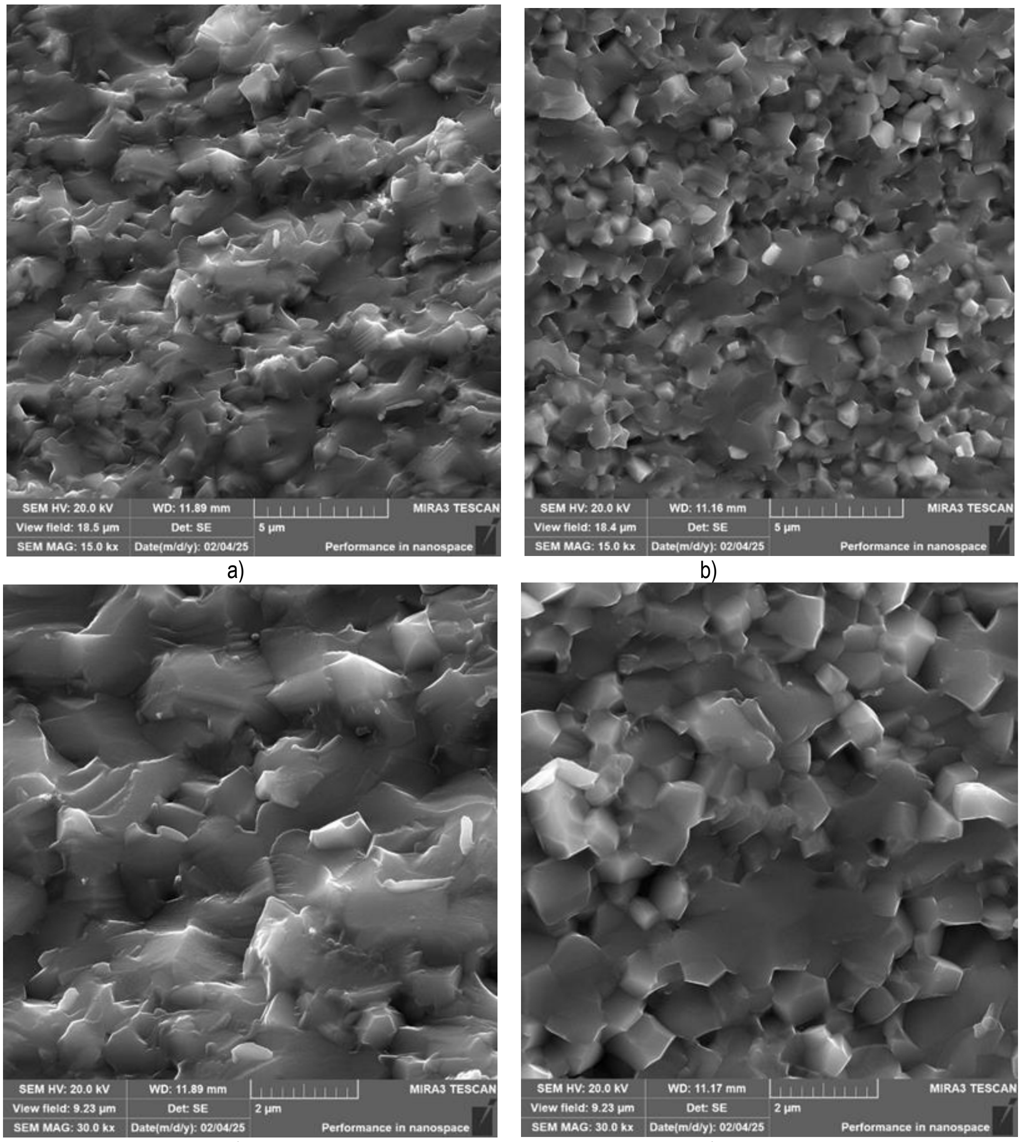


Figure 7 – Particle size distribution of powder after ultrasonic mechanoactivation



a) b)

Figure 8 – Pictures of the initial powder (a) and after ultrasonic mechanoactivation (b)



c) d)

Figure 9 – Microstructure of the sintered press blank obtained without the application of ultrasound (a, c) and obtained using ultrasound (b, d)

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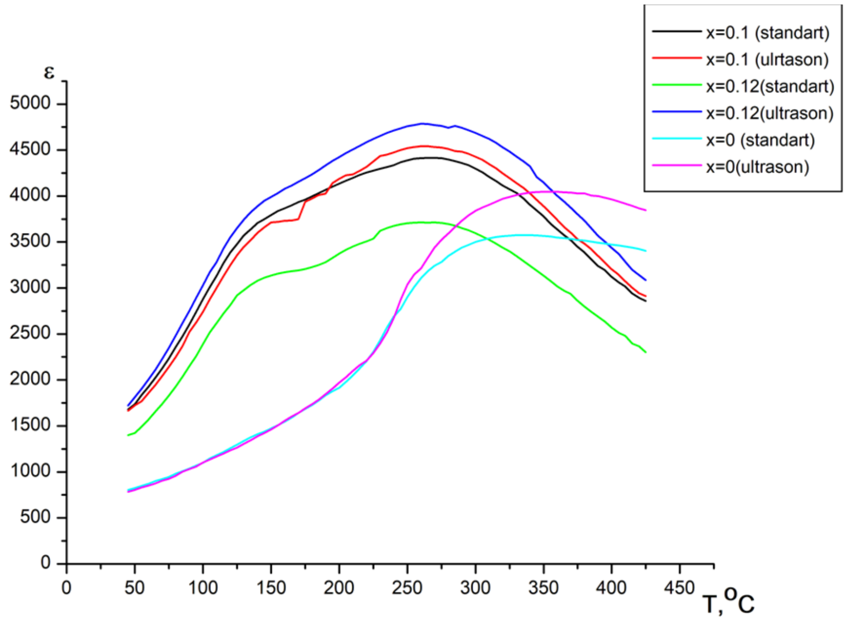

Figure 10 – Temperature dependence of dielectric constant ε

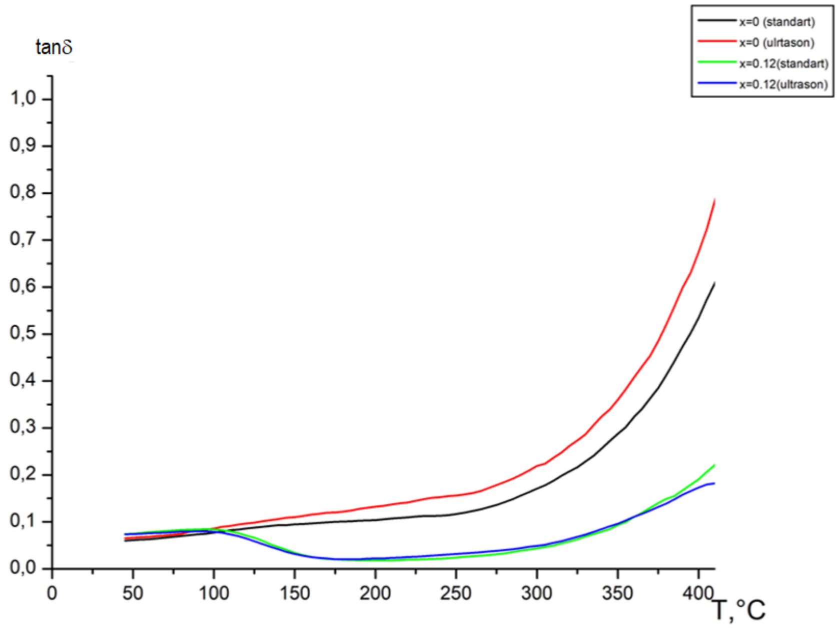

Figure 11 – Temperature dependence of the dielectric loss tangent

Conclusion

The application of ultrasonic vibrations during mechanoactivation and pressing of the synthesized powder of NBT-based piezoceramics provides the obtaining of denser homogeneous ceramics, preventing the formation of microcracks during sintering, reducing the sintering temperature by ~60 °C, increasing the Curie temperature by 50–60 °C, and increasing the dielectric constant. Ceramics produced with ultrasound is characterized by high electromechanical anisotropy (d33/d31), which is important for practical applications when it is necessary to excite certain vibration modes. The obtained results will serve as a basis for further development of the scientific field of ultrasonic mechanoactivation and pressing of piezoceramic powders.

The research project was supported by the Belarusian Republican Foundation for Fundamental Research grants Т23КУБ-023 and PN223LH010-083. The research done in the University of Aveiro was supported by the project CICECO-Aveiro Institute of Materials, UIDB/50011/2020, UIDP/50011/2020 & LA/P/0006/2020, financed by national funds through the FCT/MEC (PIDDAC).

References

1. Ul'trazvukovaya tekhnologiya izgotovleniya konstrukcionnoj i funkcional'noj nanokeramiki / O. L. Hasanov, V. M. Sokolov, E. S. Dvilis, YU. P. Poholkov // Perspektivnye materialy. – 2002. – № 1. – C. 76–83.
2. Hasanov, O. L. Nauchnye osnovy suhogo kompaktirovaniya ul'tradispersionnyh poroshkov v tekhnologii izgotovleniya nanokeramiki : diss. … doktor tekhnich. nauk : 05.17.11 / Hasanov Oleg Leonidovich ; Tomskij politekhnicheskij universitet. – Tomsk, 2003. – 371 s.
3. Pressovanie poroshkov titanata bariya s nalozheniem ul'trazvukovyh kolebanij / V. V. Klubovich, V. K. Vysockij, G. S. Bobrovskaya [i dr.] // Izv. ANB. ser fiz-tekhn. – 1994. – № 3. – C. 16–19.
4. Pugachev, S. I. Fizicheskie i tekhnologicheskie zadachi ul'trazvukovogo formoobrazovaniya izdelij iz p'ezokeramicheskih poroshkov: tezisy dokladov regional'noj NTK / S. I. Pugachev, D. O. Haritonov. – SPb. : SPbGMTU. – 1998. – S. 40–41.
5. Ul'trazvukovoe formoobrazovanie v tekhnologicheskom processe izgotovleniya izdelij iz nemetallicheskih poroshkov : trudy 3-j Mezhdunar. konf. po morskim intellektual'nym tekhnologiyam "Morintekh-99" / M. A. Krasavina, F. F. Legusha, S. I. Pugachev, D. O. Haritonov. – SPb., 1999. – T. 1. – C. 235–237.

6. Ul'trazvukovoe formoobrazovanie izdelij iz p'ezokeramicheskih poroshkov : trudy 3-j Mezhdunar. nauch.-tekhn. konf. "APEP-96" / N. M. Popov, S. I. Pugachev, N. G. Semenova, D. O. Haritonov. – Novosibirsk, 1996. – T. 1. – C. 192.
7. Il'yushchenko, A. F. Poluchenie vysokoplotnyh keramik razlichnogo naznacheniya s ispol'zovaniem nanorazmernyh poroshkov i energii vzryva. / A. F. Il'yushchenko // Nanostrukturnye materialy – 2008: Belarus' – Rossiya – Ukraina : materialy Pervoj mezhdunar. nauch. konf., 22–25 aprelya 2008 g., g. Minsk / NAN Belarusi, Ross. AN, NAN Ukrainy [i dr.]. – Minsk, 2008. – S. 345–347.
8. Effekty moshchnogo ul'trazvukovogo vozdejstviya na strukturu i svojstva nanomaterialov : ucheb. posobie / O. L. Hasanov, E. S. Dvilis, V. V. Polisadova, A. P. Zykova. – Tomsk : Tomskij politekhnich. un-t, 2008. – 149 s.
9. SHilin, A. D. Ul'trazvukovaya mekhanoaktivaciya poroshkovyh materialov / A. D. SHilin, V. V. Rubanik, V. V. Rubanik – ml. // Perspektivnye materialy i tekhnologii ; pod red. V. V. Klubovicha. – Vitebsk : VGTU, 2013. – Gl. 30. – S. 630–651.
10. Effect of High-Power Ultrasound Treatment on Microstructure, Dielectric and Piezoelectric Properties of Complex Perovskite Oxides / A. D. Shilin, V. V. Rubanik, V. V. Rubanik – jr. [et al.] // 20th International Conference on Diffusion in Solids and Liquids, Barcelona, June 24–28, 2024. – Barcelona, 2024.
11. Ul'trazvukovaya mekhanoaktivaciya poroshka p'ezokeramiki Na0.5Bi0.5TiO3 / V. V. Rubanik, V. K. Frolov, N. S. Kolodinskaya, A. Atvars // Aktual'nye problemy prochnosti : materialy nauch.-praktich. konf., g. Vitebsk, 23–27 maya 2022 g. / pod red. V. V. Rubanika. – Minsk : IVC Minfina, 2022. – S. 406–408.
12. Klubovich, V. V. Vliyanie ul'trazvukovyh kolebanij na process uplotneniya poroshkovyh materialov / V. V. Klubovich, V. V. Rubanik // Aktual'nye problemy prochnosti : materialy 43-j Mezhdunar. konf., Vitebsk, 27 sent. – 1 okt. 2004 g. : v 2 ch. / VGTU. – Vitebsk, 2004. – CH. 1. – S. 325–329.
13. Pat. 2007272 Rossijskaya Federaciya, MKI5 V22F 3/02. Sposob pressovaniya poroshkovogo materiala : № 4892743/02 : zayavleno 19.12.90 : opubl. 15.02.94 / Klubovich V. V., Bobrovskaya G. S., Rubanik V. V., Vysockij V. K. : zayavitel' Vitebskoe otdlel. in-ta fiziki tverdogo tela i poluprovodnikov AN Belarusi // Byul. Izobreteniya. – 1994. – № 3. – S. 87.
14. Patent BY 22360, MPK B 22F 3/02 (2006.01), B 30B 11/14 (2006.01). Sposob pressovaniya poroshkovogo materiala : № 20170007 : zayavleno 09.01.2017 : opubl. 30.08.2018 / Rubanik V. V., Rubanik V. V. (ml.), SHilin A. D., Salak A., Don Din Fung, chan Bao chung ; zayavitel': Institut tekhnicheskoj akustiki NAN Belarusi. – 4 s.
15. Structural and dielectric features of (Bi0.5Na0.5)1−xBaxTiO3 lead-free ferroelectric ceramics: An approach to the phase diagram / B. R. Moya, A. C. Iglesias-Jaime, A. C. Silva [et al.] // J. Appl. Phys. – 2024. – Vol. 135. – Art. 164106. – DOI: 10.1063/5.0191402.
16. Ferroelectric domain switching pathways – From grain boundary to grain body / X. Lou, X. Hou, H. Fan [et al.] // Appl. Phys. Lett. – 2023. – Vol. 122. – Art. 212902. – DOI: 10.1063/5.0146252.
17. Understanding the grain size dependence of functionalities in lead-free (Ba,Ca)(Zr,Ti)O3 / M.-H. Zhanga, Ch. Zhao, X. Yan [et al.] // Acta Mater. – 2024. – Vol. 276. – Art. 120112. – DOI: 10.1016/j.actamat.2024.120112.
18. Grain size effect on piezoelectric properties of rhombohedral lead zirconate titanate ceramics / Zh. Li, Ch.-B.-W. Li, H.-Ch. Thong [et al.] // Ceram. Inter. – 2023. – Art. 27733–27741. – DOI: 10.1016/j.ceramint.2023.05.209.
19. Bokov, A. A. Recent Progress in Relaxor Ferroelectrics with Perovskite Structure / A. A. Bokov, Z.-G. Ye // Prog. Adv. Dielectr. – 2020. – P. 105–164. – DOI: 10.1007/s10853-005-5915-7.
20. Cross, L. E. Relaxor ferroelectrics / L. E. Cross // Ferroelectrics. – 1987. – Vol. 76. – P. 241–267. – DOI: 10.1080/00150198708016945.

Список цитированных источников

1. Ультразвуковая технология изготовления конструкционной и функциональной нанокерамики / О. Л. Хасанов, В. М. Соколов, Э. С. Двилис, Ю. П. Похолков // Перспективные материалы. – 2002. – № 1. – С. 76–83.
2. Хасанов, О. Л. Научные основы сухого компактирования ультрадисперсионных порошков в технологии изготовления нанокерамики : дисс. … доктор технич. наук : 05.17.11 / Хасанов Олег Леонидович ; Томский политехнический университет. – Томск, 2003. – 371 с.
3. Прессование порошков титаната бария с наложением ультразвуковых колебаний / В. В. Клубович, В. К. Высоцкий, Г. С. Бобровская [и др.] // Изв. АНБ. сер физ-техн. – 1994. – № 3. – С. 16–19.
4. Пугачев, С. И. Физические и технологические задачи ультразвукового формообразования изделий из пьезокерамических порошков: тезисы докладов региональной НТК / С. И. Пугачев, Д. О. Харитонов. – СПб. : СПбГМТУ. – 1998. – С. 40–41.
5. Ультразвуковое формообразование в технологическом процессе изготовления изделий из неметаллических порошков : труды 3-й Междунар. конф. по морским интеллектуальным технологиям "Моринтех-99" / М. А. Красавина, Ф. Ф. Легуша, С. И. Пугачев, Д. О. Харитонов. – СПб., 1999. – Т. 1. – С. 235–237.
6. Ультразвуковое формообразование изделий из пьезокерамических порошков : труды 3-й Междунар. науч.-техн. конф. "АПЭП-96" / Н. М. Попов, С. И. Пугачев, Н. Г. Семенова, Д. О. Харитонов. – Новосибирск, 1996. – Т. 1. – С. 192.
7. Ильющенко, А. Ф. Получение высокоплотных керамик различного назначения с использованием наноразмерных порошков и энергии взрыва. / А. Ф. Ильющенко // Наноструктурные материалы – 2008: Беларусь – Россия – Украина : материалы Первой междунар. науч. конф., 22–25 апреля 2008 г., г. Минск / НАН Беларуси, Росс. АН, НАН Украины [и др.]. – Минск, 2008. – С. 345–347.
8. Эффекты мощного ультразвукового воздействия на структуру и свойства наноматериалов : учеб. пособие / О. Л. Хасанов, Э. С. Двилис, В. В. Полисадова, А. П. Зыкова. – Томск : Томский политехнич. ун-т, 2008. – 149 с.
9. Шилин, А. Д. Ультразвуковая механоактивация порошковых материалов / А. Д. Шилин, В. В. Рубаник, В. В. Рубаник – мл. // Перспективные материалы и технологии ; под ред. В. В. Клубовича. – Витебск : ВГТУ, 2013. – Гл. 30. – С. 630–651.
10. Effect of High-Power Ultrasound Treatment on Microstructure, Dielectric and Piezoelectric Properties of Complex Perovskite Oxides / A. D. Shilin, V. V. Rubanik, V. V. Rubanik – jr. [et al.] // 20th International Conference on Diffusion in Solids and Liquids, Barcelona, June 24–28, 2024. – Barcelona, 2024.
11. Ультразвуковая механоактивация порошка пьезокерамики Na0.5Bi0.5TiO3 / В. В. Рубаник, В. К. Фролов, Н. С. Колодинская, A. Atvars // Актуальные проблемы прочности : материалы науч.-практич. конф., г. Витебск, 23–27 мая 2022 г. / под ред. В. В. Рубаника. – Минск : ИВЦ Минфина, 2022. – С. 406–408.
12. Клубович, В. В. Влияние ультразвуковых колебаний на процесс уплотнения порошковых материалов / В. В. Клубович, В. В. Рубаник // Актуальные проблемы прочности : материалы 43-й Междунар. конф., Витебск, 27 сент. – 1 окт. 2004 г. : в 2 ч. / ВГТУ. – Витебск, 2004. – Ч. 1. – С. 325–329.
13. Пат. 2007272 Российская Федерация, МКИ5 B22F 3/02. Способ прессования порошкового материала : № 4892743/02 : заявлено 19.12.90 : опубл. 15.02.94 / Клубович В. В., Бобровская Г. С., Рубаник В. В., Высоцкий В. К. : заявитель Витебское отдлел. ин-та физики твердого тела и полупроводников АН Беларуси // Бюл. Изобретения. – 1994. – № 3. – С. 87.
14. Патент BY 22360, МПК B 22F 3/02 (2006.01), B 30B 11/14 (2006.01). Способ прессования порошкового материала : № 20170007 : заявлено 09.01.2017 : опубл. 30.08.2018 / Рубаник В. В., Рубаник В. В. (мл.), Шилин А. Д., Салак А., Дон Дин Фунг, чан Бао чунг ; заявитель: Институт технической акустики НАН Беларуси. – 4 с.
15. Structural and dielectric features of (Bi0.5Na0.5)1−xBaxTiO3 lead-free ferroelectric ceramics: An approach to the phase diagram / B. R. Moya, A. C. Iglesias-Jaime, A. C. Silva [et al.] // J. Appl. Phys. – 2024. – Vol. 135. – Art. 164106. – DOI: 10.1063/5.0191402.
16. Ferroelectric domain switching pathways – From grain boundary to grain body / X. Lou, X. Hou, H. Fan [et al.] // Appl. Phys. Lett. – 2023. – Vol. 122. – Art. 212902. – DOI: 10.1063/5.0146252.
17. Understanding the grain size dependence of functionalities in lead-free (Ba,Ca)(Zr,Ti)O3 / M.-H. Zhanga, Ch. Zhao, X. Yan [et al.] // Acta Mater. – 2024. – Vol. 276. – Art. 120112. – DOI: 10.1016/j.actamat.2024.120112.
18. Grain size effect on piezoelectric properties of rhombohedral lead zirconate titanate ceramics / Zh. Li, Ch.-B.-W. Li, H.-Ch. Thong [et al.] // Ceram. Inter. – 2023. – Art. 27733–27741. – DOI: 10.1016/j.ceramint.2023.05.209.
19. Bokov, A. A. Recent Progress in Relaxor Ferroelectrics with Perovskite Structure / A. A. Bokov, Z.-G. Ye // Prog. Adv. Dielectr. – 2020. – P. 105–164. – DOI: 10.1007/s10853-005-5915-7.
20. Cross, L. E. Relaxor ferroelectrics / L. E. Cross // Ferroelectrics. – 1987. – Vol. 76. – P. 241–267. – DOI: 10.1080/00150198708016945.

Material received 01.12.2025, approved 04.12.2025, accepted for publication 04.12.2025