DOI 10.1007/s10717-017-9901-8
*Glass and Ceramics*, Vol. 73, Nos. 11 – 12, March, 2017 (Russian Original, Nos. 11 – 12, November – December, 2016)

# AT ENTERPRISES AND INSTITUTES

UDC 666.76:621.386

## POSSIBILITY OF USING THE PIEZOCERAMIC PZT-19 IN PYROELECTRIC X-RAY GENERATORS

**K. A. Vokhmyanina,[1] O. O. Ivashchuk,[1] V. Yu. Ionidi,[2] A. A. Kaplii,[1] I. A. Kishchin,[1,3] A. S. Klyuev,[1] A. S. Kubankin,[1,3,5] M. V. Mishunin,[3] R. M. Nazhmudinov,[1,3] I. S. Nikulin,[1] A. N. Oleinik,[1] A. V. Sotnikov,[1] A. S. Chepurnov,[2] and A. V. Shchagin[1,4]**

Translated from *Steklo i Keramika*, No. 11, pp. 27 – 31, November, 2016.

The possibility of using PZT-19 ceramic in a pyroelectric x-ray generator is investigated experimentally. Measurements of the x-ray spectra showed the possibility of obtaining on a ceramic surface in vacuum potentials up to 7 kV, which is very low compared with typical similar values for pyroelectric crystals of lithium niobate and tantalate. This feature is due to the significant permittivity of the ceramic. It is shown that the main criterion for picking a ceramic for a pyroelectric x-ray generation could be the maximum value of the ratio of the pyroelectric constant to the permittivity.

***Key words:*** piezoelectric ceramic, pyroelectric effect, x-ray generator.

The possibility of generating x-rays to 100 keV by using pyroelectric materials has been known for quite a long time [1] and is now being actively studied [2 – 5]. This possibility is due to the pyroelectric effect, where an electric field appears in pyroelectric materials as a result of a change in their temperature. Under vacuum conditions with residual gas pressure of the order of $1.33 \times 10^{-7}$ MPa ($10^{-3}$ Torr) the electric charge induced on the surface of the pyroelectric is not shielded from the surrounding environment, as a result of which an electric field with intensity of the order of $10^5 – 10^6$ V/m arises near the surface of the pyroelectric. This electric field can be used to generate x-rays by placing a grounded target opposite the surface of the pyroelectric material, whose temperature varies cyclically. Depending on the phase of the temperature change the direction of the field reverses, as a result of which the direction of motion of the particles (electrons and ions) changes. It is obvious that particles accelerated in this manner can generate x-rays.

As a rule, in pyroelectric x-ray generators pyroelectric single crystals of lithium niobate $LiNbO_3$ and lithium tantalate $LiTaO_3$ are used as the pyroelectric material [6]. Their use made it possible to obtain experimentally x-rays with energies up to 350 keV [7].

Recently, however, interest has appeared in piezoelectric ceramics, which also possess pyroelectric properties, for example, ceramics in the lead zirconate titanate family $Pb(Zr_xTi_{1-x})O_3$ (PZT). Polydomain polycrystalline samples of PZT are widely used in ferroelectric cathodes to obtain high-current electron beams [5]. One of the first works on x-ray generation using pyroelectric ceramics [8] showed promise in using the piezoelectric ceramic zirconate-titanate lead borate TsTBS-3M in a pyroelectric source, which made it possible to obtain x-ray spectra up to 65 keV with integral radiation yield comparable to that for pyroelectric generators based on single crystals.

In the present work we investigate the possibility of using PZT-19 ceramic in a pyroelectric x-ray generator. This ceramic is one of the most common ceramics of this type produced on a commercial scale and it is inexpensive. In this work x-rays were generated by varying the temperature of PZT-19, and an electric current flowing from the surface of

[1] International Scientific and Educational Laboratory of Radiation Physics, Belgorod State University, Belgorod, Russia.
[2] D. V. Skobel'tsyn Scientific-Research Institute of Nuclear Physics, Moscow State University, Moscow, Russia.
[3] P. N. Lebedev Physical Institute, Russian Academy of Sciences, Moscow, Russia.
[4] National Scientific Center – Khar'kov Physico-technical Institute, Khar'kov, Ukraine.
[5] E-mail: kubankin@bsu.edu.ru.

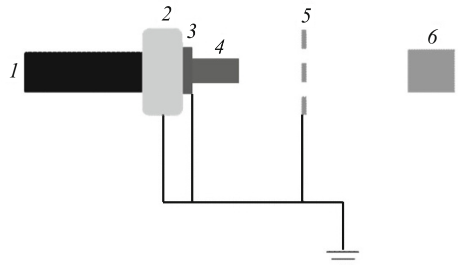


**Fig. 1.** Experimental setup for measuring the x-ray spectrum of the pyroelectric generator with piezoceramic PZT-19: *1* ) vacuum manipulator; *2* ) radiator; *3* ) Peltier element; *4* ) PZT-19 piezoelectric ceramic; *5* ) target (brass mesh); *6* ) semiconductor x-ray detector radiation.

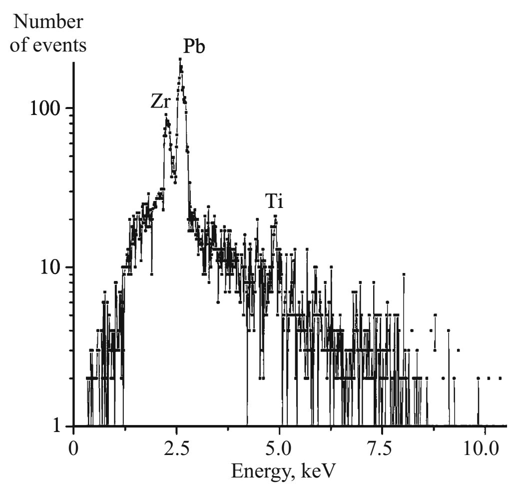


**Fig. 2.** Integral spectrum of a pyroelectric x-ray source with piezoelectric PZT-19.

the piezoelectric ceramic upon a change in temperature was measured. The data made it possible to determine the pyroelectric constant of PZT-19, compare it with the data for the pyroelectric constant for single crystals, and draw a conclusion about the impact of the permittivity of the pyroelectric material on the capacity of the pyroelectric to be an effective x-ray generator.

## EXPERIMENT

This work was conducted at the International Scientific and Educational Laboratory of Radiation Physics at Belgorod State University. The measurements of the x-ray spectrum from the pyroelectric generator with the piezoceramic PZT-19 were performed on the experimental setup shown in Fig. 1. The pyroelectric x-ray generator (PXG) included a sample of the piezoelectric ceramic PZT-19 secured to a Peltier element, which, in turn, was mounted on a duraluminum radiator. All parts of the sources are connected to each other by a conducting epoxy adhesive. The PZT-19 sample

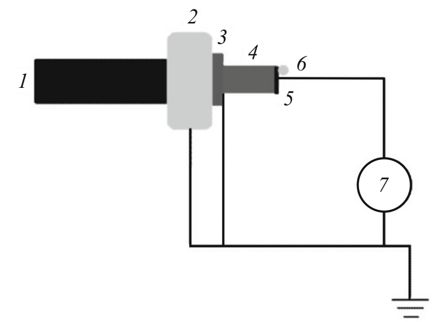


**Fig. 3.** Experimental arrangement for measuring the current from the surface of the piezoceramic PZT-10: *1* ) vacuum manipulator; *2* ) wall of vacuum chamber; *3* ) radiator; *4* ) Peltier element; *5* ) piezoceramic PZT-19; *6* ) DS18B20 heat sensor; *7* ) Keithley 6485 picoammeter.

comprised a 10 mm in diameter and 15 mm high cylinder. The PXG was placed inside a vacuum chamber and could be moved along the axis of the PXG by using a vacuum manipulator.

The radiator and the surface of the Peltier element, on which the piezoelectric ceramic sample was placed, were grounded. A grounded brass mesh with width 0.2 mm, which was used as a target, was placed 10 mm from the working surface of PZT-19. A semiconductor x-ray detector was placed about 200 mm from the working surface of the crystal.

Ten cycles of heating and cooling of the piezoceramic in the range 20 – 45°C at a residual gas pressure about $1.33 \times 10^{-7}$ MPa ($10^{-3}$ Torr) were conducted. The total spectrum obtained upon heating and cooling is displayed in Fig. 2. In all 7950 photons were detected in 1800 sec. The majority of the events were recorded at positive potential of the surface of the piezoelectric ceramics (corresponding to heating). The evidence for this is the presence of several characteristic lines of the elements making up the lead titanate-zirconate: zirconium Zr line ($L_\alpha$-line) — 2.043 keV, lead Pb line ($M_\alpha$-line) — 2.345 keV, titanium Ti line ($K_\alpha$-line) — 4.511 keV. It should be noted that there are no copper or zinc lines (elements present in the target), which is explained by that fact that the energy of the accelerated electrons is insufficient to excite the characteristic lines.

On the whole the intensity and energy values for the radiation from the generator with the piezoceramic PZT-19 turned out to be very low and not comparable to the parameters of the spectra of pyroelectric sources with lithium tantalate and lithium niobate and TSTBS piezoceramic. To make the pyroelectric properties more precise the current and charge of the working surface piezoceramic were measured as a function of the surface temperature. This measurement makes it possible to compare the pyroelectric constant in different pyroelectric materials.

The experimental arrangement for measuring the current and charge from the surface of the piezoelectric PZT-19 is shown in Fig. 3. A metallic electrode, completely covering

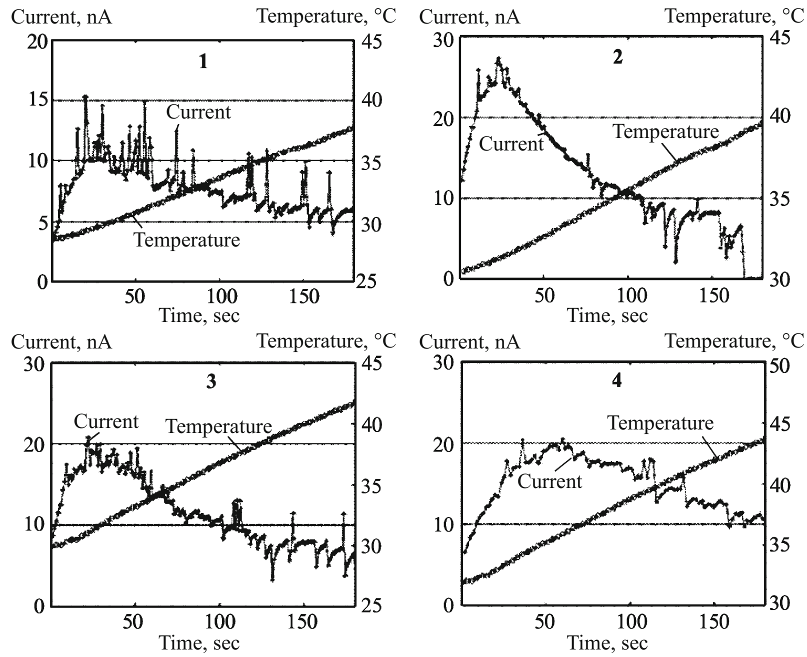


**Fig. 4.** Measurements (1 – 4) of the current from the surface of the ceramic and the temperature upon heating the ceramic PZT-19: the cycle numbers correspond to Table 1.

the surface of the ceramic, was secured on the working surface of the ceramic with the aid of conducting epoxy adhesive.

The current from the electrode was measured using a picoammeter. A Peltier element was used to change the temperature of the ceramic; a TST isolated from the measuring circuit was used to monitor the temperature. The measurements were conducted under residual gas pressure $1.33 \times 10^{-6}$ MPa ($10^{-2}$ Torr).

The measurements of the current from the surface of the ceramic and the temperature are presented in Fig. 4 for heating of the piezoceramic and Fig. 5 for cooling. The data for four successive cycles are shown.

Since the measurement was performed from a surface that is perpendicular to the pyroelectric axis, the experimentally measured current is predominantly of piezoelectric origin. Since no external force was applied to the ceramic, the piezoelectric effect did not manifest and did not contribute to the current measurements. The measurements showed that the characteristic value of the current from the PZT-19 surface is comparable to that of the current from the surface of a lithium niobate crystal in a comparable temperature range [9]. Knowing the temperature change and the total charge as well as the surface area of the piezoceramic the piezoelectric constant of the experimental sample can be determined from the relation

$$p = \frac{QS}{\Delta T}, \qquad (1)$$

where $p$ is the pyroelectric constant, $C/(K \times cm^{2)})$; $Q$ is the integral charge, C; $S$ is the area of the ceramic, $cm^2$; and, $\Delta T$ is the temperature range, °C.

The computational results are presented in Table 1.

The computed value of the pyroelectric constant fluctuates from $3.373 \times 10^{-8}$ $C/(K \times cm^2)$ to $1.191 \times 10^{-7}$ $C/(K \times cm^2)$

**TABLE 1.** Determination of the Pyroelectric Constant from the Experimental Data

| Order of measurement | Total charge (in modulus), C | Temperature change over measurement time, °C (K) | Working surface area of piezoceramic, cm$^2$ | Pyroelectric constant, $C/(K \times cm^2)$ |
|---|---|---|---|---|
| Heating 1 | $1.409 \times 10^{-6}$ | 11.38 | 0.785 | $9.723 \times 10^{-8}$ |
| Heating 2 | $2.086 \times 10^{-6}$ | 15.19 | 0.785 | $1.078 \times 10^{-7}$ |
| Heating 3 | $2.285 \times 10^{-6}$ | 15.69 | 0.785 | $1.143 \times 10^{-7}$ |
| Heating 4 | $1.617 \times 10^{-6}$ | 11.88 | 0.785 | $1.191 \times 10^{-7}$ |
| Cooling 1 | $1.085 \times 10^{-6}$ | 11.25 | 0.785 | $7.570 \times 10^{-8}$ |
| Cooling 2 | $1.512 \times 10^{-6}$ | 12.00 | 0.785 | $9.891 \times 10^{-8}$ |
| Cooling 3 | $4.727 \times 10^{-7}$ | 11.00 | 0.785 | $3.373 \times 10^{-8}$ |
| Cooling 4 | $1.030 \times 10^{-6}$ | 11.75 | 0.785 | $6.881 \times 10^{-8}$ |

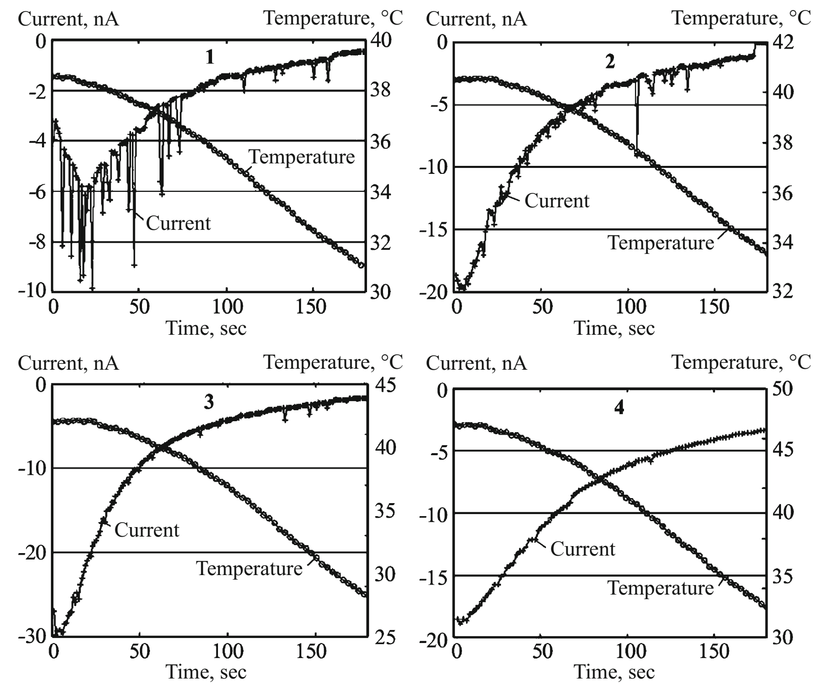


**Fig. 5.** Measurements (1 – 4) of the current from the surface of the ceramic and the temperature upon cooling of the ceramic PZT-19: the cycle numbers correspond to Table 1.

with average value $9.379 \times 10^{-8} \pm 1.20 \times 10^{-8}$ C/(K × cm$^2$). Comparing this value with the average value of the pyroelectric constant for lithium niobate $4 \times 10^{-9}$ C/(K × cm$^2$) [6] we find that the pyroelectric constant of PZT-19 is an order of magnitude higher than that of lithium niobate. This fact is completely at variance with the measurements of the x-ray yield from a pyroelectric generator on PZT-19. Thus, the pyroelectric constant is not the primary characteristic of the capacity to produce a strong external electric field.

A comparative analysis of the PZT-19 and lithium niobate parameters showed that the discrepancy between the experimental measurements of the pyroelectric current and the data on the x-ray spectrum could be due to the significant difference in the permittivity å of both materials. It is evident that the permittivity of PZT-19 ($\varepsilon = 1350$ according to the manufacturer) is almost two orders of magnitude higher than that of lithium niobate ($\varepsilon = 30$ according to the data in [6]). Since the permittivity of a medium characterizes the factor by which the electric field in the medium is lower than in vacuum, it becomes clear why in comparing the amount of charge generated in the pyroelectric effect the electric field is weaker in the case of PZT-19 and, correspondingly, the x-ray spectrum is also weaker.

Comparing the ratio of pyroelectric constant to the permittivity for lithium niobate and PZT-19 the following values obtain. The ratio $p/\varepsilon = 1.32 \times 10^{-10}$ for lithium niobate and $p/\varepsilon = 6.94 \times 10^{-11}$ for PZT-19. The ratio of the pyroelectric constant to the dielectric constant of PZT-19 is thereby lower than in lithium niobate. The physical meaning of this ratio determines a part of the charge generated by the pyroelectric effect and creating an external electric field relative to the pyroelectric. Apparently, a more informative and characteristic quantity for determining the applicability of a pyroelectric material in a pyroelectric generator is precisely the ratio of the pyroelectric constant to the permittivity.

## CONCLUSIONS

The possibility of using the piezoelectric ceramic PZT-19 in a pyroelectric x-ray generator was investigated. The experimental results were contradictory. On the one hand, in the case of the pyroelectric effect a quite high current is recorded from the PZT-19 surface, indicating a higher value of the pyroelectric constant than in the case of the lithium niobate and tantalate single-crystals conventionally used in pyroelectric generators. On the other hand the x-ray spectrum from a source with PZT-19 ceramic is quite low-energy bounded by about 9 keV.

Analysis of the parameters of the pyroelectric materials used in pyroelectric sources shows that the permittivity has a considerable effect. The ratio of the pyroelectric constant to the permittivity can serve as a more reliable indicator for evaluating the applicability of a pyroelectric material in a pyroelectric x-ray generator.

In summary, it was shown that PZT-19 ceramic cannot be a prospective material for a pyroelectric accelerator and the main criterion for selecting pyroelectric materials for a pyroelectric accelerator could be the maximum value of the ratio of pyroelectric constant to the dielectric constant.

*This work was supported under a program of the Ministry of Education and Science of the Russian Federation at institutions of higher learning, contract No. 14.578.21.0192 (RFMEFI57816X0192).*

## REFERENCES


1. J. D. Brownridge, "Pyroelectric x-ray generator," *Nature*, **358**, 277 – 278 (1992).
2. J. D. Brownridge and S. M. Shafroth, "Self-focused electron beams produced by pyroelectric crystals on heating or cooling in dilute gases," *Appl. Phys. Lett.*, **79**, 3364 – 3366 (2001).
3. J. Geuther, Y. Danon, F. Saglime, and B. Sones, "Electron acceleration for x-ray production using paired pyroelectric crystals," in: *Abstracts of the 6th International Meeting on Nuclear Applications of Accelerator Technology [AccApp'03]*, San Diego, CA, June 1 – 5, 2003, San Diego (2003), pp. 591 – 594.
4. O. O. Ivashuk, A. S. Kubankin, A. N. Oleinik, and A. V. Shchagyn, "Investigation of the x-ray yield from pyroelectric sources with conical targets," *Pov-st', Rentgen., Sinkhrotr., Neutron. Issled.*, No. 8, 1 – 5 (2016).
5. G. Rosenman, D. Shur, Ya. E. Krasik, and A. Dunaevsky, "Electron emission from ferroelectrics," *J. Appl. Phys.*, **88**, 6109 – 6161 (2000).
6. K. K. Wong, "Properties of lithium niobate," in: *INSPEC*, The Institution of Electrical Engineers, London (2002), pp. 200 – 208.
7. W. Tornow, S. M. Lynam, and S. M. Shafroth, "Substantial increase in acceleration potential of pyroelectric crystal," *J. App. Phys*., **107**, 063302 (2010).
8. A. V. Shchagin, V. S. Miroshnik, V. I. Volkov, and A. N. Oleinik, "Ferroelectric ceramics in a pyroelectric accelerator," *Appl. Phys. Lett*., **107**, 233505 (2015).
9. J. D. Brownridge and S. M. Shafroth, *New Research on Lasers and Electrooptics*, Nova Science Publishers, New York (2007).