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Piezoceramic resistance to radiation amorphization during operation in ITER

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Received April 1, 2022
Revised October 5, 2022
Accepted October 8, 2022

Piezoelectric motors designed for operation in the ITER tokamak-reactor must be tested for stability under severe radiation conditions. Properties of lead zirconate-titanate that is the most common type of piezoelectric materials were analyzed from the point of view on influence of radiation. It is shown that, at the expected in ITER level of radiation, this piezoceramics has a good potential for resistance to radiation induced amorphization and depolarization.

Keywords: piezoceramics, radiation tests, ITER diagnostics.

DOI: 10.21883/TPL.2022.12.54935.19208

International experimental fusion reactor ITER will be exploited under extremely high temperatures and densities of fusion plasma. To ensure its reliable operation, it is necessary to apply diagnostic equipment for controlling plasma temperature, pressure, magnetic fields, radiation energy and density, as well as other performance parameters. The efficiency and reliability of the diagnostic equipment operation under severe radiation conditions in the fusion reactor are important factors of the thermonuclear program. Numerous diagnostic mirrors need protection against erosion and deposition of materials during the fusion plasma disruptions. One of the basic components of mirror protection will be several tens of shutters [1] to be driven by mechanisms that will operate in vacuum under high heat and neutron fluxes being maintained remotely as rare as once in 20 years. Such operating conditions impose distinctive requirements both for materials and devices based on these materials. Segnetoelectrics and piezoelectric ceramics based on them are regarded as promising materials for fusion reactors because their properties are resistant to high levels of radiation [2]. It is known that significant variations in properties of piezoceramic materials are observed only when integral fast−neutron fluence exceed 10^{17} n/cm^2 (> 0.1 MeV). Figs. 1, *a* and *b* present the results of the neutron−physics analysis of neutron and gamma flux distributions in the ITER's lower diagnostic port [3]. The arrow points at locations of the piezomotors under development [4] installed at less than 1 m from fusion plasma. As the figure shows, at this point the neutron and gamma fluence of $10^{19}\,\mathrm{cm}^{-2}$ ($E > 0.1$ MeV) is ~ 50 times higher than the radiation dose collected during testing motors designed by the PI Company which were assumed to be used in better−protected ITER areas [5]. The radiation background was calculated using the MCNP code for 2D electron temperature and density distributions under ITER baseline inductive burning plasma conditions at $Q = 10$.

The piezoceramics properties change under irradiation due to the impact of neutrons and gamma photons. The neutron effects may be subdivided into, first, elastic collisions with nuclei which cause energy transfer to the nuclei according to the law of solid−sphere collisions and, second, nucleus reactions whose products induce ionization of the medium and elastic displacements of atoms. Direct elastic displacements can generate only neutrons with energies of > 0.1 MeV. Vice versa, nucleus reaction cross−sections are especially large in the range of low neutron energies ($\ll 0.1$ MeV). Interaction of gamma photons with matter may proceed through the elastic scattering channel either without energy losses and Compton scattering accompanied by arising of fast recoil electrons, or with absorption accompanied by electron emission, or with generating electron−positron pairs at the gamma−photon energies of > 1 MeV. Photons with energies of > 10 MeV may interact with nuclei with emission of a proton, neutron, or α-particle and recoil nucleus. In this work we have estimated the effect of transmutation, gas generation and other radiation−induced processes on the lead zirconate−titanate piezoceramics $PbZr_{1-x}Ti_xO_3$ (PZT). The calculation was performed for the PZT composition $0.5PbTiO_3{-}0.5PbZrO_3$ taking into account the fact that the maximum piezoeffect is observed for solid solutions located near the morphotropic phase boundary at the basic−components concentration $x \sim 0.5$ [6]. Various radiation−induced processes were calculated using the FISPACT-II program code and data on cross−sections of the relevant processes [7–10] (Tables 1 and 2). The calculated parameters presented in the tables are accurate to the third decimal place; the data shown in the tables are given with accounting for the possible stoichiometric deviation of up to the second decimal place. Code FISPACT-II supports the calculation of results of elastic and inelastic radiation−induced reactions; in addition, it considers such channels of neutron−induced reactions

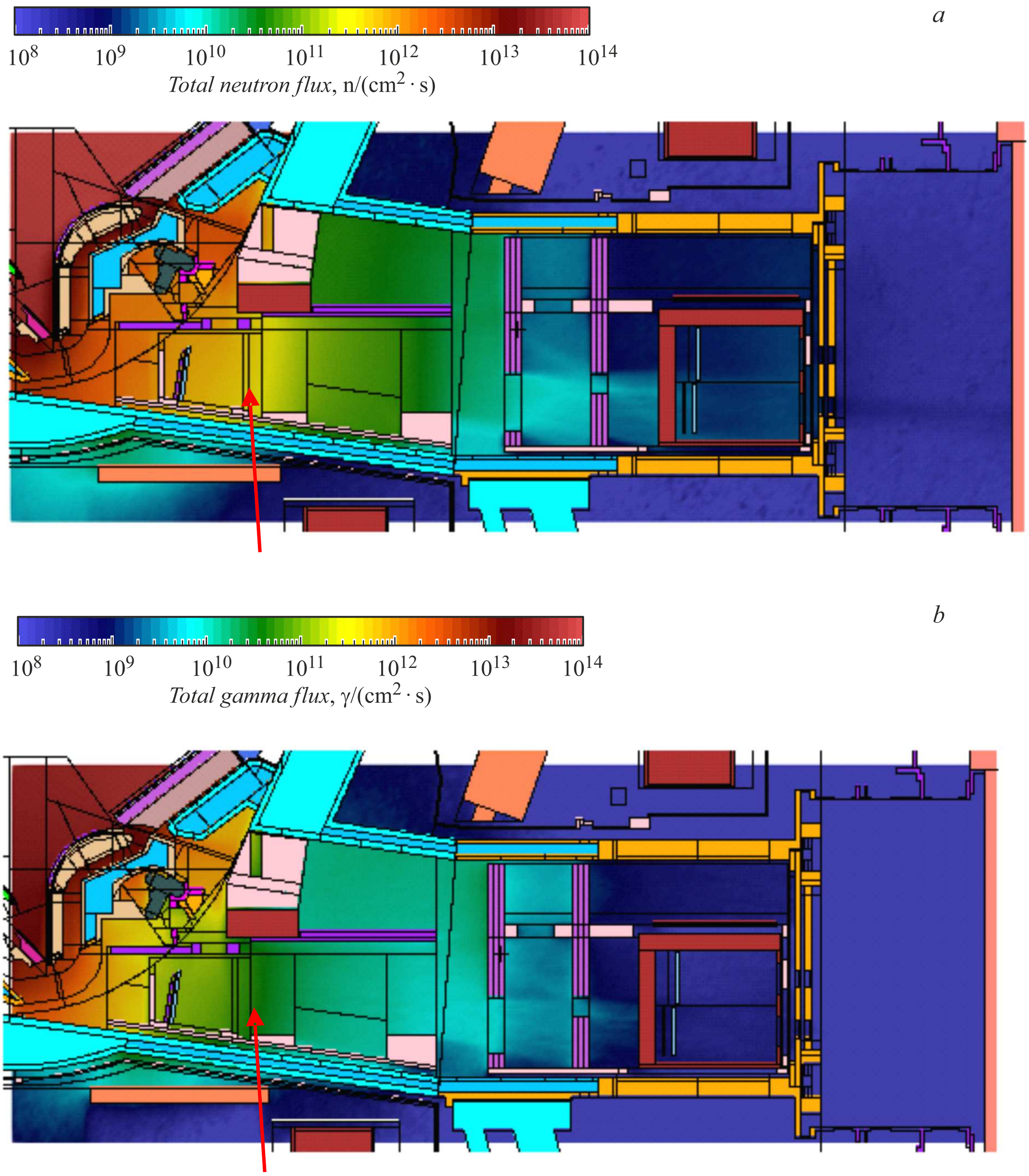

Figure 1. Vertical cross−section of the MCNP model of divertor port # 8 with diagnostic equipment. *a* — neutron fluxes, *b* — gamma ray fluxes.

where there are emitted two neutrons $(n, 2n)$, neutron and α-particle $(n, n\alpha)$, neutron and proton (n, np), two neutrons and α-particle $(n, 2n\alpha)$, or neutron and two α-particles $(n, n2\alpha)$. Tables 1 and 2 demonstrate relative influences of inelastic processes, gas generation and atomic displacement. One can see that the probability of gas generation is million times lower. The obtained results showed that the most probable reactions among the entire variety of analyzed ones are reactions resulting in displacement of atoms from their lattice sites. Displacement of $\sim 2\%$ of all the atoms may result in amorphization (destruction) of the PZT crystal structure and, hence, in reduction or total loss of the piezoeffect [11]. Fig. 2 presents the possible phases of an individual cell of the perovskite ABO_3 crystal: cubic (*a*), tetragonal (*b*), monoclinic (orthorhombic) extended along the face diagonal (*c*), and rhombohedral extended along the volume diagonal (*d*). The PZT piezoceramic materials are known to be solid solutions located near the mor-

Table 1. Estimation of the number of atomic displacements in the PZT ceramics

Reaction type	$F^{*}_{n\text{-}tot}(>0.1\,\mathrm{MeV}) \sim 2.25 \cdot 10^{12}\,\mathrm{n/(cm^2 \cdot s)}$		Fluence$^{**}(>0.1\,\mathrm{MeV}) \sim 3.8 \cdot 10^{19}$ n/cm^2
	Displacements per second per 1 g	DPA***/s	DPA*** (total)
(n, D), decay	$1.30 \cdot 10^{11}$	$1.40 \cdot 10^{-11}$	$2.4 \cdot 10^{-4}$
(n, D), inelastic	$4.68 \cdot 10^{11}$	$5.04 \cdot 10^{-11}$	$8.5 \cdot 10^{-4}$
(n, D), elastic	$1.01 \cdot 10^{13}$	$1.08 \cdot 10^{-9}$	$1.9 \cdot 10^{-2}$
(n, D), total	$1.10 \cdot 10^{13}$	$1.19 \cdot 10^{-9}$	$2.0 \cdot 10^{-2}$

* In 14 years of ITER operation.
** Total fluence.
*** DPA — displacements per atom.

Table 2. Gaseous products per 1 g of the PZT ceramics ($F_{n\text{-}tot} = 2.25 \cdot 10^{12}\,\mathrm{n/(cm^2 \cdot s)}$) in 14 years of ITER operation at the total fluence of $\sim 3.8 \cdot 10^{19}\,\mathrm{n/cm^2}$)

Element	Post−irradiation time		
	0	1 year	10 years
	appm*		
He-4	0.243	0.243	0.243
He-3	$2.3 \cdot 10^{-6}$	$2.7 \cdot 10^{-6}$	$5.1 \cdot 10^{-6}$
H-3	$6.5 \cdot 10^{-6}$	$6.1 \cdot 10^{-6}$	$3.7 \cdot 10^{-6}$
H-2	0.011	0.011	0.011
H-1	0.050	0.050	0.050

*appm — atomic parts per million.

photropic phase boundary with coexistence of the tetragonal (Fig. 2, *b*) and rhombohedral (Fig. 2, *d*) phases [12]. It was assumed that commercially available compositions with the maximum tetragonal deformation (ratio between linear sizes along the axes *c* and *a*) which belong to this solid−solution group will exhibit higher radiation resistance of the crystal lattices.

Paper [13] showed that irradiation of the perovskite–structure materials causes expansion of the crystal lattice independently of their dielectric properties. In cubic−lattice materials that do not exhibit piezoelectric properties this expansion is isotropic and linearly increases with increasing dose. Expansion in noncentrosymmetric perovskites with different *c* and *a* parameters is of a more complex character. In the process of radiation−induced lattice expansion of some perovskite−structure materials, lattice parameter *a* increases faster than *c*, while tetragonality c/a decreases continuously thus making the material closer to the symmetric cubic phase having no piezoelectric properties. However, there exist compounds, e. g., $PbTiO_3$ (one of the main components of PZT piezoceramics) where anisotropy of the radiation−induced expansion is such that the radiation dose increase first leads to an increase in tetragonality which means that the lattice structure does not get closer to the cubic one but deviates from it [13]. Paper [14] showed also that tetragonality of the $PbTiO_3$ structure increased with the radiation impact increasing to fluence ($> 0.1\,\mathrm{MeV}$) of $\sim 10^{20}\,\mathrm{n/cm^2}$ (at the irradiated samples temperature of 40−70°C). The presented data were taken as a base for considering the radiation resistance of the PZT piezoceramics itself.

Neutron and gamma−photon impacts on the material structure and, hence, on the piezoelectric properties, are of different natures. The effect of gamma photons reduces actually to formation of fast secondary electrons in the following elementary processes: Compton scattering accompanied by formation of fast recoil electrons and photoelectron absorption. The main changes induced by gamma photons concern the crystal electronic structure and are, to high extent, of the dynamic character. Fast neutron fluxes ($E > 0.1\,\mathrm{MeV}$) can damage the crystal structure much more severely than gamma rays because the energy they transfer in elastic collisions with crystal lattice atoms is ~ 1000 times higher. Knocked−out heavy particles decelerate in the matter due to losing energy in exciting and ionizing the atoms, as well as in elastic collisions with nuclei. Thus, damages due to elastic interactions and radiation−induced amorphization depend mainly on the fast−neutron fluence ($E > 0.1\,\mathrm{MeV}$) [2]. The state of initial damage is not, certainly, the final result of the material radiation damage. After the initial damage has arisen, the formed defects become mobile and can annihilate, create larger clusters of defects or enlarge the amorphous zone sizes, disappear on the surface or grain boundaries, give rise to dislocations, enlarge dislocations, promote the dislocation displacements, etc. [15]. It is known that amorphization does not occur at a sufficiently high temperature, i. e., the perovskite structure is able to self−recover; for a number of perovskites, including titanates to which $PbTiO_3$ belongs, this temperature limit is as low as 100−200°C [16]. Jointly with the analysis of available literature data, the consideration performed in this work has shown that the PZT piezoceramics has a good potential for resistance to irradiation up to the fluence of $\sim 10^{19}$−$10^{20}\,\mathrm{n/cm^2}$ ($> 0.1\,\mathrm{MeV}$). At the same time, radiation−induced amorphization and depolarization of the PZT ceramics under the conditions similar to those

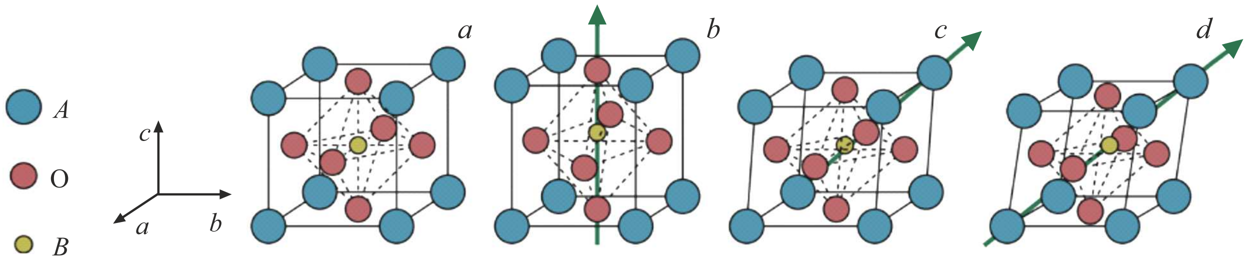

Figure 2. Individual crystal cell of perovskite ABO_3. (*a*) — cubic phase, (*b*) — tetragonal phase, (*c*) — monoclinic (orthorhombic) phase with extension along the face diagonal, and (*d*) — rhombohedral phase with extension along the volume diagonal.

at ITER have been studied only fragmentarily and need more investigation. The problem is not restricted to studying only deterioration of piezoelectric properties and needs experimental investigation of the possibility to retain the ceramics performance characteristics and restore its serviceability under the condition of radiation fluxes and periodic heating expected to occur at ITER; in addition, it is necessary to demonstrate its operation within piezoelectric motors.

Financial support

The study was partially funded in the framework of the Ioffe Institute State Assignment.

Conflict of interests

The authors declare that they have no conflict of interests.

References

[1] C. Vorpahl, A. Alekseev, S. Arshad, T. Hatae, A. Khodakhi, J. Klabachahi, F. Le Guern, E. Mukhin, S. Pak, C. Seon, M. Smith, E. Yatsuka, A. Zvonkov, Fusion Eng. Des., **123** (11), 712 (2017). DOI: 10.1016/j.fusengdes.2017.05.111

[2] M.G. Cain, P.M. Weaver, M.J. Reece, J. Mater. Chem. A, **4** (27), 10394 (2016). DOI: 10.1039/C6TA01935H

[3] E.E. Mukhin, V.M. Nelyubov, V.A. Yukish, E.P. Smirnova, V.A. Solovei, N.K. Kalinina, V.G. Nagaitsev, M.F. Valishin, A.R. Belozerova, S.A. Enin, A.A. Borisov, N.A. Deryabina, V.I. Khripunov, D.V. Portnov, N.A. Babinov, D.V. Dokhtarenko, I.A. Khodunov, V.N. Klimov, A.G. Razdobarin, S.E. Alexandrov, D.I. Elets, A.N. Bazhenov, I.M. Bukreev, An.P. Chernakov, A.M. Dmitriev, Y.G. Ibragimova, A.N. Koval, G.S. Kurskiev, A.E. Litvinov, K.O. Nikolaenko, D.S. Samsonov, V.A. Senichenkov, R.S. Smirnov, S.Yu. Tolstyakov, I.B. Tereschenko, L.A. Varshavchik, N.S. Zhiltsov, A.N. Mokeev, P.V. Chernakov, P. Andrew, M. Kempenaars, Fusion Eng. Des., **176** (9), 113017 (2022). DOI: 10.1016/j.fusengdes.2022.113017

[4] E. Mukhin, P. Andrew, N. Babinov, M. Bassan, A. Bazhenov, I. Bukreev, Al. Chernakov, An. Chernakov, A. Dmitriev, V. Yukish, M. Kochergin, A. Koval, G. Kurskiev, A. Litvinov, V. Nelyubov, A. Razdobarin, D. Samsonov, V. Semenov, V. Solokha, V. Solovey, S. Tolstyakov, M. Walsh, Fusion Eng. Des., **123** (11), 686 (2017).
DOI: 10.1016/j.fusengdes.2017.06.014

[5] M. Pillon, Ch. Monti, G. Mugnaini, C. Neri, P. Rossi, M. Carta, O. Fiorani, A. Santagata, Fusion Eng. Des., **96-97**, 329 (2015). DOI: 10.1016/j.fusengdes.2015.03.025

[6] G.A. Smolensky, V.A. Bokov, V.A. Isupov, N.N. Kraynik, R.E. Pasynkov, A.I. Sokolov, N.K. Yushin, *Fizika segnetoelektricheskikh yavleniy* (Nauka, L., 1985). (Inn Russian)

[7] R.A. Forrest, J. Kopecky, J.Ch. Sublet, *The European activation file: EAF-2007 neutron-induced cross section library* (EURATOM/UKAEA Fusion Association, Culham Science Centre, Abingdon, U.K., 2007), UKAEA FUS 535.

[8] J.-Ch. Sublet, J.W. Eastwood, J.G. Morgan, M.R. Gilbert, M. Fleming, W. Arter, Nucl. Data Sheets, **139**, 77 (2017). DOI: 10.1016/j.nds.2017.01.002

[9] A.J. Koning, D. Rochman, J. Ch. Sublet, *TENDL-2017: TALYS-based evaluated nuclear data library* [Electronic source] (release date: December 30, 2017).
https://tendl.web.psi.ch

[10] M.J. Loughlin, N.P. Taylor, *ITER Report ITER_D_2V3V8G* (2009), vol. 1.1.

[11] E.V. Peshikov, *Deystvie radiatsii na segnetoelektriki* (Fan, Tashkent, 1972). (In Russian)

[12] F.D. Bloss, *Crystallography and crystal chemistry* (Mineralogical Society of America, Washington, DC, 1994).

[13] O. Hauser, M. Schenk, Phys. Status Solidi B, **18** (2), 547 (1966). DOI: 10.1002/pssb.19660180208

[14] S.P. Solov'ev, V.Ya. Dudarev, V.V. Zakurkin, I.I. Kuz'min, Izv. AN SSSR. Ser. fiz., **XXXV** (9), 1931 (1971). (in Russian)

[15] K. Nordlund, S.J. Zinkle, A.E. Sand, F. Granberg, R.S. Averback, R.E. Stoller, T. Suzudo, L. Malerba, F. Banhart, W.J. Weber, F. Willaime, S.L. Dudarev, D. Simeone, J. Nucl. Mater., **512**, 450 (2018). DOI: 10.1016/j.jnucmat.2018.10.027

[16] A. Meldrum, L.A. Boatner, W.J. Weber, R.C. Ewing, J. Nucl. Mater., **300** (2-3), 242 (2002).
DOI: 10.1016/S0022-3115(01)00733-4