

INFLUENCE OF HIGH-ENERGY ELECTRONS IRRADIATION ON NANOCERAMICS PROPERTIES OF ZIRCONIA

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Radiation defects in nanoceramics zirconia were investigated after electron irradiation with energy 41...89 MeV up to dose $2.28...5.7 \cdot 10^{14}$ electrons/cm². Radioactive isotopes ^{87,88}Y, ^{88,89,95}Zr, ⁹⁵Nb, ¹⁷⁵Hf are registered after irradiation of electrons with energy 41 and 47.2 MeV. The activity of ⁸³Rb was registered after irradiation by electrons with energy 86 and 88.9 MeV. Activity of ⁸³Rb can be formed in reactions (γ , 6n) or (e, 6n). Different process of annealing radiation defects in samples of nanoceramics is detected after irradiated by electrons with energy 41...47.2 or 86...88.9 MeV. The colour centres for samples of nanoceramics after irradiated by electrons with energy 41...47.2 MeV practically completely was annealed at 500°C. For samples of nanoceramics after irradiated by electrons with energy 86...88.9 MeV and after annealing at 500°C significant absorption for lengths of waves of 430 and 470 nm is observed. It is supposed that these distinctions at annealing the colour centres of zirconia are caused by formation in nuclear reactions by high-energy electrons on isotopes of a matrix of krypton atoms. The krypton atoms are the centres segregation point defects. As a result the steady defective complexes were formed.

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INTRODUCTION

Dielectric resonator for wakefield accelerators should have high values of dielectric constant ($\epsilon > 20$) and small dielectric losses ($\text{tg}\delta < 10^{-4}$) and to keep these parameters during acceleration of particles. Therefore influence of radiation damages on dielectric parameters flying relativistic electron bunches represents significant interest [1, 2].

One of possible materials for dielectric waveguides can be zirconia. It has good mechanical and ionic properties. It is used as a grinding medium and engineering ceramics. Therefore, the material properties, inducing optical properties, are of a high interest. The favourable properties of ZrO₂-based materials become especially pronounced if these materials were fabricated of powders of nanoscopic size. It is well known that the phase stabilization of zirconia can be achieved by an addition of a certain amount of the trivalent yttrium oxide (Y₂O₃, yttria) to form a solid solution in the ZrO₂ lattice.

At operation of the dielectric accelerator there will be an activation of resonators due to losses of an accelerating beam. Research of activity of dielectric resonators which were irradiated by relativistic electrons is of interest.

The purpose of the present work is research of formation of radioactive elements and radiation defects in nanoceramic ZrO₂(Y₂O₃) at an irradiation by relativistic electrons.

RESULTS AND DISCUSSION

ZrO₂ – 3 mol% Y₂O₃ nanopowders (3Y-TZP) were synthesized by a co-precipitation technique using ZrOCl₂·nH₂O salts. Calcinations temperature of dried zirconium hydroxides was chosen at 700°C with dwelling time 2 hours. This calcinations temperature provides the formation of unagglomerated nanopowders and sin-

tering obtained nanopowders to density near theoretical at 1400...1500°C. The density was 6.0...6.02 g/cm³ for ceramic materials, obtained by sintering of zirconia nanopowders obtained from ZrOCl₂·nH₂O.

Radiation defects in nanoceramics zirconia were investigated after electron irradiation with energy 41...89 MeV up to a dose $2.28...5.7 \cdot 10^{14}$ electrons/cm² (Figs. 1, 2).

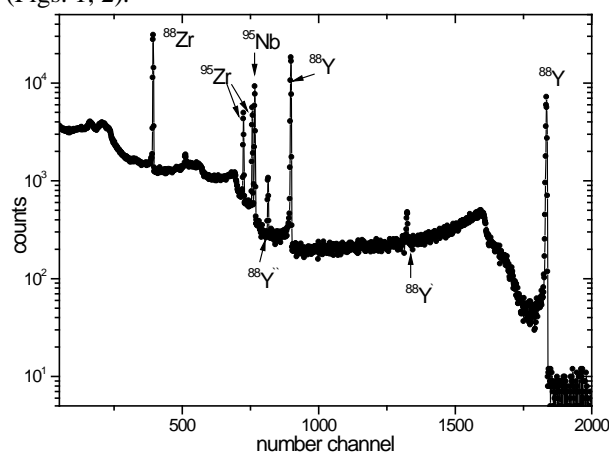


Fig. 1. The spectrum of nanoceramics after irradiation at 47.2 MeV by dose $2.28 \cdot 10^{14}$ electrons/cm²

Activity of the isotopes generated by electron irradiation of samples of nanoceramics was measured by Ge(Li)-detector. Radioactive isotopes ^{87,88}Y, ^{88,89,95}Zr, ⁹⁵Nb, ¹⁷⁵Hf are registered after irradiation of electrons with energy 41 and 47.2 MeV (see Fig. 1). At an irradiation by electrons and bremsstrahlung of nanoceramics in Zr of natural isotopic content (⁹⁰Zr...51.46%; ⁹¹Zr...11.23%; ⁹²Zr...17.11%; ⁹⁴Zr...17.40%; ⁹⁶Zr...2.8%) the highest activity is realized from isotope ⁸⁹Zr, received in reaction ⁹⁰Zr(γ ,n)⁸⁹Zr (Fig. 3) [3].

The half-life period of ⁸⁹Zr equals to 79.3 hours and in long-term aspect it does not represent danger. Also activity and from an isotope ⁹⁵Zr from reaction

$^{96}\text{Zr}(\gamma, n)^{95}\text{Zr}$ with a half-life period of 64.05 days can be realized.

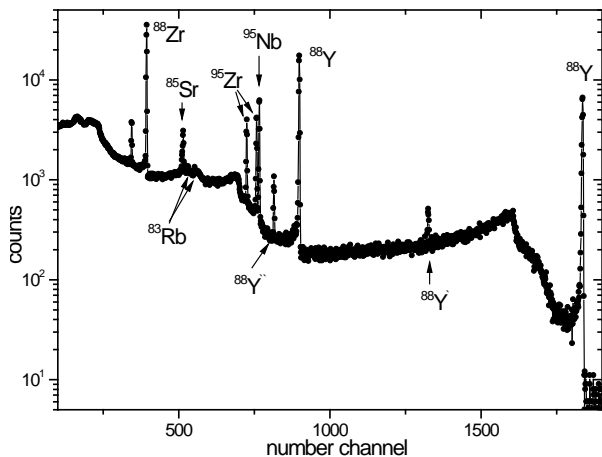


Fig. 2. The spectrum of nanoceramics after irradiation at 86 MeV by dose $5.7 \cdot 10^{14}$ electrons/cm²

However on an isotope ^{94}Zr can activity from ^{93}Zr with $T_{1/2}=9.5 \cdot 10^5$ years is realized. During work of the ceramic resonator its irradiation by bremsstrahlung and neutrons, arising in accompanying reactions, can be realized. For example, ^{93}Zr can arise in reaction of radiation capture on ^{92}Zr . Therefore for significant activity of ^{93}Zr it is necessary to carry out its burial in geological structures. From the point of view of radiating safety at operation of wake-field accelerators the greatest danger represents ^{88}Y which is formed in reactions $^{89}\text{Y}(\gamma, n)^{88}\text{Y}$ or $^{89}\text{Y}(e, n)^{88}\text{Y}$. The half-life period ^{88}Y amounts 106.65 days. Disintegration ^{88}Y is accompanied by radiation of two intensive lines with energy 898 and 1836 keV.

The activity of ^{83}Rb was registered after irradiation by electrons with energy 86 and 88.9 MeV (see Fig. 2). Activity of ^{83}Rb can be formed in reactions $(\gamma, 6n)$ or $(e, 6n)$. The threshold of the given reactions equals to 59.4 MeV. The activity of ^{88}Zr is determined by reaction $^{90}\text{Zr}(\gamma, 2n)^{88}\text{Zr}$ (Fig. 4). Therefore the ^{88}Zr activity of nanoceramic is noticeably more for irradiation by electrons with energy 86 MeV.

Kerma-constants Γ_δ of radionuclide's and power of the air kerma $\Gamma_\delta \cdot A(\text{Bk})$ on distance of 1 m at the moment of an irradiation (normalization on $m=1$ g)

Nu- c- leus	$T_{1/2}$ (days)	γ -rays, E_γ , keV (n_i %)	Γ_δ , aGy·m ² /s·Bk	$\Gamma_\delta \cdot A(\text{Bk})$ ($D=2.3E+14$ e ⁻ /cm ² , E=47.2 MeV)	$\Gamma_\delta \cdot A(\text{Bk})$ ($D=4.9E+14$ e ⁻ /cm ² , E=41 MeV)	$\Gamma_\delta \cdot A(\text{Bk})$ ($D=5.7E+14$ e ⁻ /cm ² , E=86 MeV)	$\Gamma_\delta \cdot A(\text{Bk})$ ($D=5.4E+14$ e ⁻ /cm ² , E=88.9 MeV)
1	2	3	4	5	6	7	8
^{88}Zr	83.4	392.9 (100)	12.99	$2.16 \cdot 10^5$	$3.65 \cdot 10^5$	$5.25 \cdot 10^5$	$7.40 \cdot 10^5$
^{89}Zr	3.27	909.0 (99.87)	30.05	$1.53 \cdot 10^8$	—	—	—
^{95}Zr	64.02	724.4 (44.15) 756.7 (54.46)	27.12	$3.66 \cdot 10^5$	$6.72 \cdot 10^5$	$6.28 \cdot 10^5$	$9.18 \cdot 10^5$
^{95}Nb	34.98	765.8 (99.81)	27.97	$2.29 \cdot 10^5$	$1.21 \cdot 10^6$	$9.44 \cdot 10^5$	$1.39 \cdot 10^6$
^{87}Y	3.33	388.5 (82.10) 484.8 (89.74)	24.9	$3.10 \cdot 10^6$	—	—	—
^{88}Y	106.65	898.0 (93.68), 1836 (99.24)	88.05	$2.56 \cdot 10^6$	$4.72 \cdot 10^6$	$5.66 \cdot 10^6$	$7.56 \cdot 10^6$
^{175}Hf	70.	343.4 (84.0)	9.54	$1.53 \cdot 10^4$	—	$3.05 \cdot 10^4$	—
^{93}Rb	—	—	—	—	—	14.9	22.92

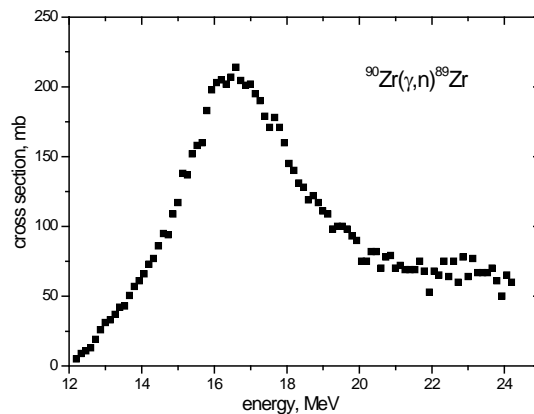


Fig. 3. Cross section $^{90}\text{Zr}(\gamma, n)^{89}\text{Zr}$

Dependence of activity nanoceramics from time with yttria and without yttria is resulted on Fig. 5.

Kerma-constants Γ_δ of radionuclides and power of air kerma $\Gamma_\delta \cdot A(\text{Bk})$ on distance of 1 m at the moment of an irradiation were shown in table. Isotopes with small time half-decay do not represent danger at operation of the dielectric accelerator. It is possible to see, that the greatest danger at operation of dielectric accelerators is represented by isotopes ^{88}Zr , ^{95}Zr , ^{95}Nb and ^{88}Y which have big times of half-decay.

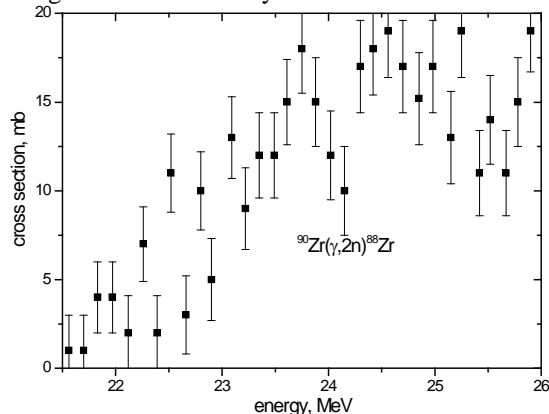


Fig. 4. Cross section $^{90}\text{Zr}(\gamma, 2n)^{88}\text{Zr}$

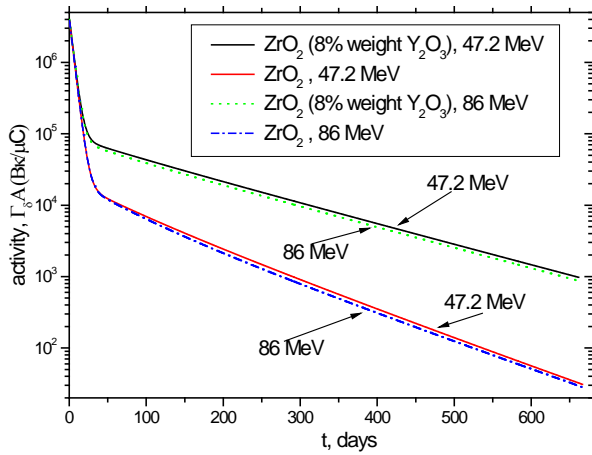


Fig. 5. Activity nanoceramics from time with yttria and without yttria

The maximal energy of recoil atoms E_{rec} of oxygen or zirconium in nanoceramics at irradiation by relativistic electrons equals:

$$E_{rec} = \frac{2E_1(E_1 + 2m_0c^2)}{M_2c^2}, \quad (1)$$

where m_0 , E_1 – weight and energy electrons, M_2 – weight of atoms of oxygen or zirconium. For energy electrons 47.2 MeV the maximal energy of recoil atoms of oxygen will amount 302 keV. For atoms of recoil zirconium the maximal energy of recoil atoms will amount 53 keV. For energy electrons 86 MeV the maximal energy of recoil atoms of oxygen will amount 998 keV. For atoms of recoil zirconium the maximal energy of recoil atoms will amount 175 keV. The number of the displaced atoms at braking recoil atoms with such energy can amount 6000, 1000 (47.2 MeV) and $\approx 2 \cdot 10^4$, 3500 (86 MeV), respectively. Recoil atoms with such energy can cause thermal spike. It in turn can result in transformation of initial structure of zirconia. Cross section of such processes is insignificantly. Average energy of primary displacement atoms is equaled:

$$\bar{E} = \{(E_d \cdot E_{max}) / (E_{max} - E_d)\} \ln(E_{max} / E_d), \quad (2)$$

where E_d – threshold displacement energy, E_{max} – maximum of energy of recoil atoms. Suppose that for zirconium and oxygen $E_d = 50$ eV, the average energy of primary displacement atoms will equal for oxygen 453 and 495 eV for energy electrons 47.2 and 86 MeV, respectively, and for Zr 348 and 408 eV for energy electrons 47.2 and 86 MeV, respectively.

UV-VIS spectroscopy was used for measurement of spectra of absorption of the irradiated samples of zirconia nanoceramics before and after annealing during 2 hours at 500°C. Measurements of absorption spectra carried out concerning unirradiated sample of nanoceramics. Up to annealing samples of zirconia nanoceramics the wide peak of absorption in area of 500...700 nm is observed. Also are registered absorption lines of radiation at 402.2 and 635 nm which correspond to the F and F'-centres of monocline lattices of zirconia (Figs. 6, 7) [4].

Different process of annealing radiation defects in samples of zirconia nanoceramics is detected after irradiated by electrons with energy 41...47.2 or 86...88.9 MeV.

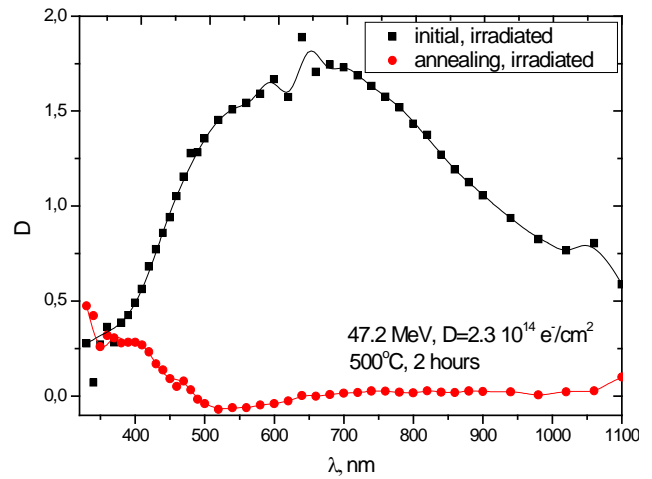


Fig. 6. UV-VIS spectra of zirconia samples of nanoceramics irradiated by relativistic electrons with energy 47.2 MeV before and after annealing

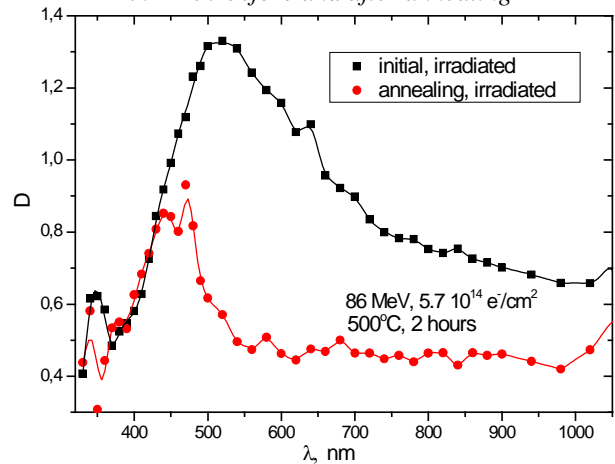


Fig. 7. UV-VIS spectra of zirconia samples of nanoceramics irradiated by relativistic electrons with energy 86 MeV before and after annealing

The color centres for samples of zirconia nanoceramics after irradiated by electrons with energy 41...47.2 MeV practically completely was annealed at 500°C (see Fig. 6). For samples of zirconia nanoceramics after irradiated by electrons with energy 86...88.9 MeV and after annealing at 500°C significant absorption for lengths of waves of 430 and 470 nm is observed (see Fig. 7). It is supposed that these distinctions at annealing the colour centres of zirconia are caused by formation in nuclear reactions by high-energy electrons on isotopes of a matrix of krypton atoms. The krypton atoms are the centers segregation point defects. As a result the steady defective complexes were formed [5 - 8].

CONCLUSIONS

1. Formation of radioactive isotopes is investigated at irradiation relativistic electrons with energy up to 100 MeV. The present data are necessary at planning a choice of a material of a dielectric wave guide of wake-field accelerators. The greatest danger at operation of dielectric accelerators is represented by isotopes ^{88}Zr , ^{95}Zr , ^{95}Nb and ^{88}Y which have big times of half-decay.

2. Formation of radiation defects in nanoceramics is investigated. The various types of radiation defects are found out at irradiation relativistic electrons with energy

47 and 86 MeV. It is revealed, that krypton atoms are centers of segregation of point defects.

3. Formation of the F and F'-centres of monocline phases is detected at an irradiation by relativistic electrons.

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ВЛИЯНИЕ ОБЛУЧЕНИЯ ЭЛЕКТРОНАМИ ВЫСОКИХ ЭНЕРГИЙ НА СВОЙСТВА НАНОКЕРАМИКИ ДВУОКИСИ ЦИРКОНИЯ

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Исследовались радиационные дефекты в нанокерамике диоксида циркония при облучении электронами с энергией 41...89 МэВ до дозы $2,28...5,7 \cdot 10^{14}$ частиц/см². При облучении электронами с энергией 41 и 47,2 МэВ регистрируются радиоактивные изотопы ^{87,88}Y, ^{88,89,95}Zr, ⁹⁵Nb, ¹⁷⁵Hf. При облучении электронами с энергией 86 и 88,9 МэВ дополнительно регистрируется активность ⁸³Rb, которая может быть образована в реакциях (γ , bn) или (e , bn). Обнаружен различный процесс отжига радиационных дефектов в образцах нанокерамики, облученных при энергии электронов 41...47,2 и 86...88,9 МэВ. Центры окраски для образцов нанокерамики, облученных электронами с энергией 41...47,2 МэВ, практически полностью отожглись при 500 °С. Для образцов нанокерамики, облученных электронами с энергией 86...88,9 МэВ, после отжига при 500 °С наблюдается значительное поглощение для длин волн 430 и 470 нм. Предполагается, что эти различия при отжиге центров окраски двуокиси циркония обусловлены образованием в ядерных реакциях под действием высокоэнергетических электронов на изотопах матрицы атомов криптона, которые являются центрами сегрегации точечных дефектов, что приводит к образованию устойчивых дефектных комплексов.

ВПЛИВ ОПРОМІНЕННЯ ЕЛЕКТРОНАМИ ВИСОКИХ ЕНЕРГІЙ НА ВЛАСТИВОСТІ НАНОКЕРАМІКИ ДВООКИСУ ЦИРКОНІЮ

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Досліджувалися радіаційні дефекти в нанокераміці двоокису цирконію при опроміненні електронами з енергією 41...89 MeV до дози $2,28...5,7 \cdot 10^{14}$ часток/см². При опроміненні електронами з енергією 41 і 47,2 MeV реєструються радиоактивні ізотопи ^{87,88}Y, ^{88,89,95}Zr, ⁹⁵Nb, ¹⁷⁵Hf. При опроміненні електронами з енергією 86 і 88,9 MeV додатково реєструється активність ⁸³Rb, що може бути утворена в реакціях (γ , bn) або (e , bn). Виявлено різний процес відпалу радіаційних дефектів у зразках нанокераміки, опроміненіх при енергії електронів 41...47,2 і 86...88,9 MeV. Центри фарбування для зразків нанокераміки, опроміненіх електронами з енергією 41...47,2 MeV, практично повністю відпалювались при 500 °С. Для зразків нанокераміки, опроміненіх електронами з енергією 86...88,9 MeV, після відпалу при 500 °С спостерігається значне поглинання для довжин хвиль 430 і 470 нм. Передбачається, що ці розходження при відпалі центрів забарвлення двоокису цирконію обумовлені утворенням у ядерних реакціях під дією високоенергетичних електронів на ізотопах матриці атомів криптона, які є центрами сегрегації крапкових дефектів, що приводить до утворення стійких дефектних комплексів.