

# Improving Structure and Superconductivity of Coated Cuprate Tapes by Irradiation with Electrons and Gamma-Rays

Shodiev Ahmad Abdunabiyevich<sup>1</sup>, Mussaeva Malika Anvarovna<sup>2</sup>, Nishonova Nodiraxon Rayimjonova<sup>3</sup>, Elmurotova Dilnoza Baxtiyorovna<sup>4</sup>, Islamova Dildora Xamidullayeva<sup>5</sup>

*1Institute of Nuclear Physics Uzbekistan Academy of Sciences, Mirzo Ulugbek district, 100214 Tashkent, Uzbekistan, \*E-mail: akhmadshodiyev@gmail.com*

*2Institute of Nuclear Physics Uzbekistan Academy of Sciences, Mirzo Ulugbek district, 100214 Tashkent, Uzbekistan, \*E-mail: mussaeva@inp.uz.*

*3Doctor of philosophy sciences, professor, head of the department "Philosophy and national idea" Tashkent State Technical University, Islam Karimov, Tashkent, University St.,*

*4 PhD, Associate Professor of the Department of Biomedical Engineering, Computer Science and Biophysics, Tashkent Medical Academy, Tashkent, Uzbekistan, E-mail: dilnoza\_elmurotova\_tma@mail.ru*

*5Candidate of philosophy of the Department of Philosophy and national idea, Tashkent State Technical University, Islam Karimov, Tashkent, University St.*

**Abstract:** The analytic data were obtained with modern techniques: XRD, M(T,H), and Hall effect (0.556 Tesla). Below the radiation damage level of destroying the superconducting state, we found such structure modifications, when magnetic flux pinning centers are generated at the concentration of  $10^{16}$ – $10^{17}$  cm<sup>-3</sup> and both  $T_c$  and  $J_c$  increase. Such an optimized current vortex state exists in 80–320 K. As irradiation with 1–5 MeV electron and 1.17–1.33 MeV gamma flux do not produce long living radio-nuclides, it is affordable for industrial technology of radiation treatment of long cable by rewinding across the flux.

**Keywords:** high- $T_c$  superconductors, electric power grid, radiation technology, pinning centers.

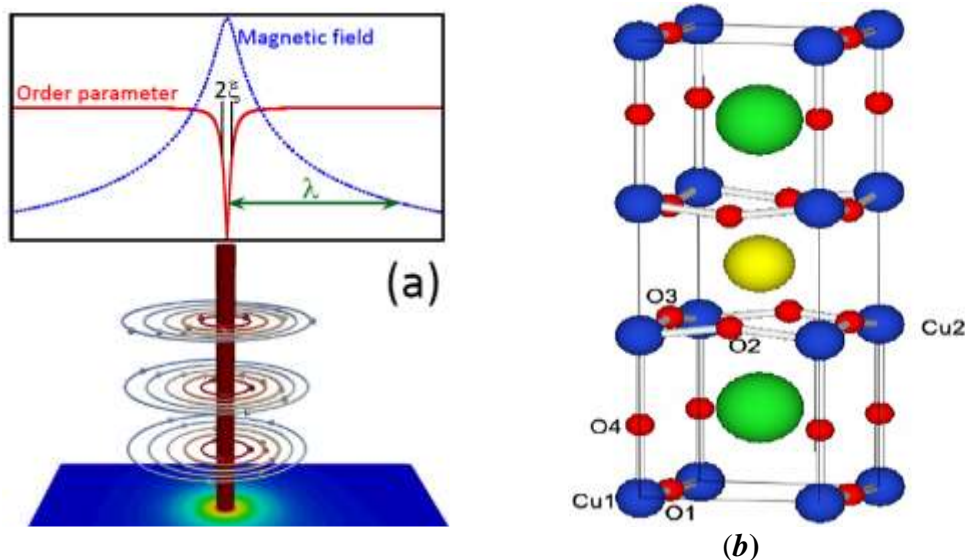
## 1. Introduction

High temperature superconductors (HTS) have been of great interest for practical applications since their discovery by Bednorz and Müller [1] with a  $T_c$  above 30 K. "Yttrium barium copper oxide" (YBCO) was the first HTS to exhibit superconductivity above the liquid nitrogen boiling point of 77 K and was discovered in 1987 by Wu [2]. It was soon followed by the discovery of superconductivity in similar materials. Its high critical temperature and its ability to withstand high magnetic fields make YBCO the superconductor of choice for future power applications such as energy transportation and generators, when fabricated in the form of wires.

Superconducting single-domain YBCO and GdBCO/Ag bulks were tested for space and nuclear applications by an exposition to increasing doses of gamma and neutron radiation in the LVR-15 research reactor [3]. Maximum doses were  $3.85 \times 10^4$  Gy from neutron and  $7.00 \times 10^5$  Gy from gamma radiation, which is much larger dose than can be received in any spacecraft during the standard space mission [3]. Recently, a new method to increase the critical current of these coated tapes at high magnetic fields with ionic irradiation was shown. The proton implantation with an energy of 4 MeV and fluence of  $8 \cdot 10^{16}$  protons/cm<sup>2</sup> twice

increased the critical currents for  $H = 6$  kOe at  $T = 27$  K [4]. Further studies showed that oxygen ions with an energy irradiation of 3.5 MeV through a protective Ag layer of a 2G HTS superconducting tape could also reproduce the doubling of the critical current density in high magnetic fields [5]. The critical temperature ( $T_c$ ) and the critical current density ( $J_c$ ) decrease as an irradiation fluence increases. It was evidenced that the superconducting phase disappears for the fluence of  $5 \cdot 10^{12}$  for  $^{132}\text{Xe}^{27+}$  ions/cm<sup>2</sup> and  $10^{13}$  for  $^{86}\text{Kr}^{17+}$  ions/cm<sup>2</sup> [6]. For protons irradiation of 2.5 MeV, the radiation resistance of the  $\text{GdBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (GdBCO) 2G HTS tape is higher than that of the YBCO tapes. Compared to the YBCO tapes, the 2G HTS tape (GdBCO-based) exhibits better irradiation resistance due to the higher density of the GdBCO than the YBCO. The threshold for a decrease of the critical current densities is  $6.2 \cdot 10^{15}$  protons/cm<sup>2</sup> for the GdBCO tapes and  $2.7 \cdot 10^{15}$  protons/cm<sup>2</sup> for YBCO tapes [6]. In 1987 the first positive results of flux pinning at strong magnetic fields, causing the magnetization and  $J_c$  enhancement after moderate doses of neutron and ion irradiations in single crystal  $\text{YBa}_2\text{Cu}_3\text{O}_{7-b}$  were published in [7]. Irradiation of YBCO single crystals with 2.5 MeV electrons in the superconducting state at 4 and 20 K to the dose  $10^{18}$  cm<sup>-2</sup> resulted in generation of positrons with a half-time 190 and 250 ps at 300 K, that is the direct evidence of nuclear reactions with Cu or O; even intensive laser pulse could induce oxygen disorder in CuO chains that recovered by heating  $> 200$  K [8].

The unit cell of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  is shown in Fig.1. (a, b) Varying the oxygen content of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  results in significant changes of its physical properties. Many studies have shown that the critical temperature and crystal structure of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  change with oxygen content [9,10]. Neutron diffraction and magnetic measurements have shown that  $T_c$  is dependent on the charge balance between the copper-oxygen chains and copper-oxygen planes [11]. The chain sites serve as charge reservoirs from which electrons are transferred to the copper-oxygen planes as the oxygen content decreases. It is within the copper-oxygen planes that superconductivity originates. As the oxygen content of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  decreases, so does  $T_c$ .



**Fig.1. (a)** Structure of an isolated vortex (red). The top graph shows the distribution of the magnetic field and order parameter amplitude in a cross-section through the vortex. The vector-lines circling the vortex represent the supercurrent screening the magnetic flux of the vortex. The bottom plane-cut shows the distribution of the order parameter. **(b)** The unit cell of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  [12].

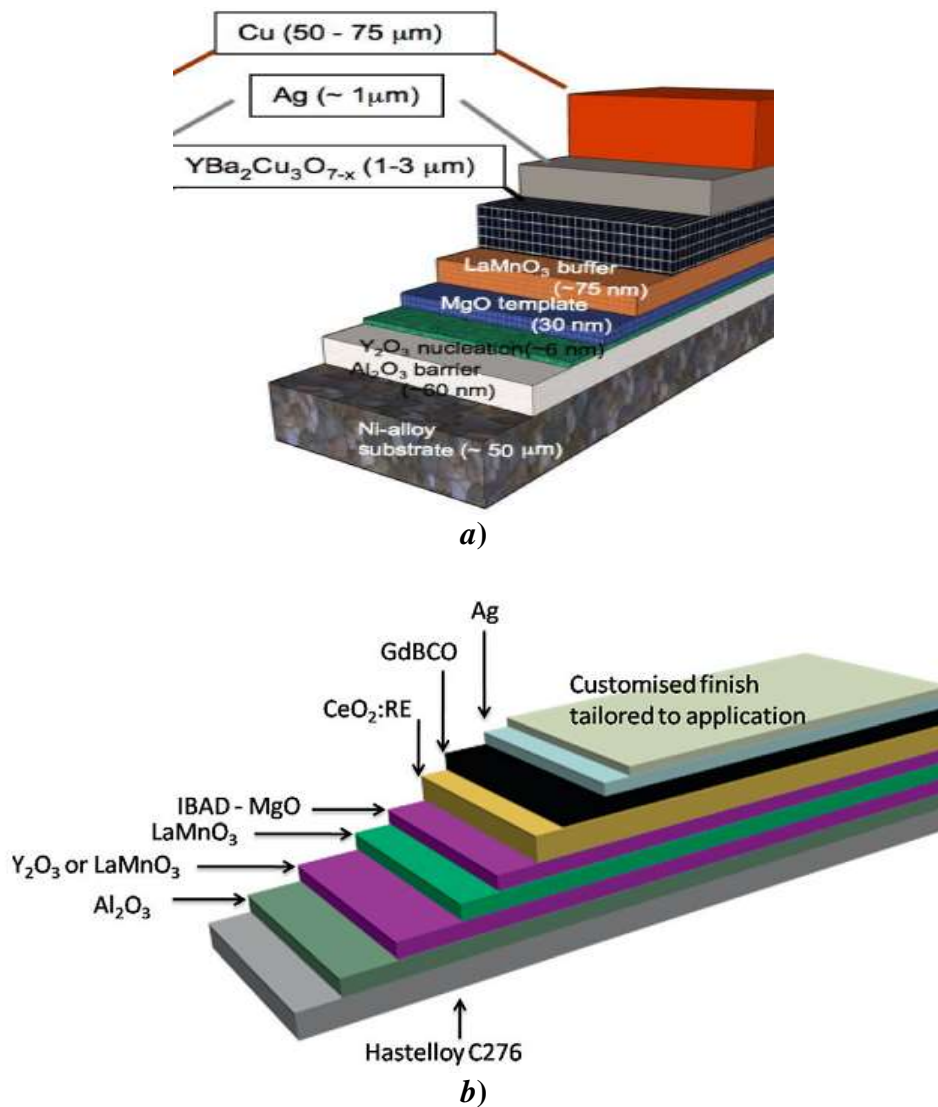


Fig. 2. Microstructure of layered coated tape  $a$ -YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub>,

$b$ - GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub> [12]

The typical structure of the YBCO tape consists of the YBCO layer, the copper stabilizer layer, the silver stabilizer layer, the Hastelloy substrate and the buffer layer which is placed between the substrate and the YBCO layer [13]. The YBCO layer, which is the only layer responsible for conducting the load current during the steady state operation, is manufactured as a film with very small thickness, protected by copper stabilizer layers on both sides [14].

HTSC-2G tapes SuperOx-YBCO-1,2 and GdBCO-3 were manufactured by S-innovation by the patented method [15]. Cables made from GdBCO tapes pass the engineer current  $> 1.5$  kA at 77 K, 3 kA at 65 K in self field, and 4 kA at 4.2 K in 8 T [16]. They contain 14-elements and have 7-layers with macroscale and micro-nanosized interfaces Ni-Cr-alloy/oxide-dielectric, HTSC/Ag, HTSC/dielectrics, Ni-alloy-Cu and Cu/Ag (fig.2). In this anisotropic structure of YBaCuO or GdBaCuO (fig.1  $b$ )  $Y^{3+}$  is a scalar nonmagnetic impurity, and  $Gd^{3+}$  is a vector magnetic impurity, each separating two adjacent  $CuO_2$  planes, where Cooper pairs move without energy dissipation [17]. Therefore their magnetic response differs one from another. Besides, the Ni- hastelloy substrate is ferromagnetic.

Here we report our latest researches on coated HTSC cuprate tapes aimed at improving crystal structure and enhancing the flux pinning at similar combined irradiation conditions.

### Samples and experimental methods.

Industrial tapes YBCO and GdBaCuO (4 mm width, 60  $\mu\text{m}$  thickness) on S-276 (Ni-Cr-Fe)-steel with a nanostructured insulating coating and covered with 3 microlayers of metals 3 $\mu\text{Ag}$ , 4 $\mu\text{Cu}$ , PbSn (trade-mark SuperOx, S-Innovations, Russia-Japan, [www.superox.ru](http://www.superox.ru)) [15,16,17,18,19]. Taking into account these results on the tapes and our early ones on YBCO crystals, we chose the following conditions: samples were irradiated in air with 5 MeV electron beam at current density 0.4–1  $\mu\text{A}/\text{cm}^2$  to doses  $10^{14}$ – $10^{15}$   $\text{el}/\text{cm}^2$  at 273–280 K, then irradiated in liquid nitrogen (77 K) by  $^{60}\text{Co}$   $\gamma$ -quanta of 1.17 and 1.33 MeV at dose rate 65  $\text{R}/\text{s}=5.85\times 10^{11}$   $/\text{cm}^2\text{s}$  in converging field geometry to doses  $< 10^6$  R, corresponding  $\sim 50$  nm spacing of  $\gamma$ - quanta, which is  $> \xi_{\text{ab}}$  and  $< \lambda_{\text{ab}}$ , and according to [20], for inducing the largest flux pinning the point defect density is  $\sim 10^{12}$   $\text{cm}^{-2}$ . Thus, these irradiation conditions would not destroy the Cooper pairs and leave some space for Abrikosov vortices around each point defect.

Crystal structure and phase composition were analyzed by X-ray diffraction spectrometer XRD Empyrean (PANalytic, Netherland).

Magneto-resistance and current carrier mobility  $\mu$  were measured by Hall method at magnetic field 0.556 Tesla  $\parallel$  c-axis in the temperature range of 80–320 K at the system HMS-7000 (Ecopia, Korea).

### Hall Effect Measurements.

Fig.3. *a,b,c* show the resistivity of the samples when  $I \parallel B$  for non-irradiated and electron irradiated samples. Doses  $10^{14}$  and  $5\cdot 10^{14}$   $\text{el}/\text{cm}^2$  were obtained at the beam current 0.4  $\mu\text{A}$ , and  $10^{15}$   $\text{cm}^{-2}$  at 1  $\mu\text{A}$ .

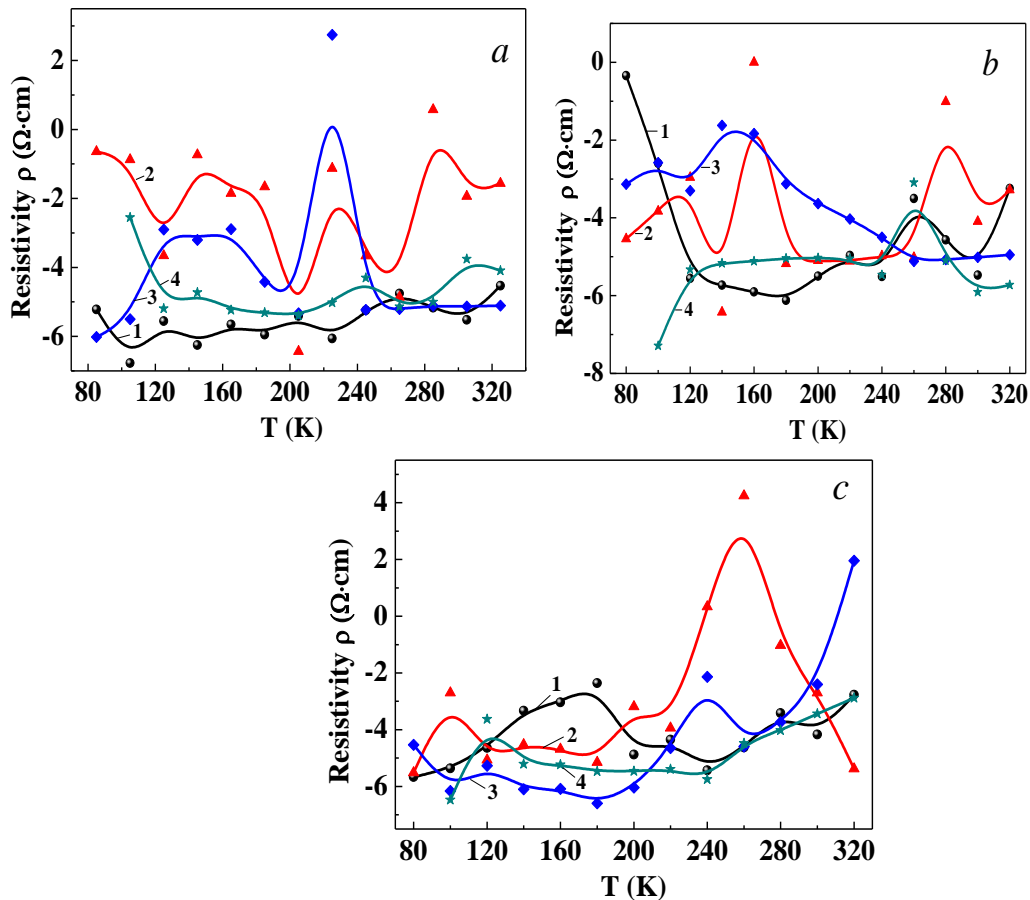


Fig. 3. Resistivity of the samples irradiated at 5 MeV electron beam in air: (a) -1-YBaCuO, (b) -2-YBaCuO, (c) -3-GdBaCuO: 1 – reference, 2 –  $10^{14}$   $\text{el}/\text{cm}^2$ ;

$3-5 \cdot 10^{14} \text{el/cm}^2$ ,  $4-10^{15} \text{el/cm}^2$

The electron dose  $10^{15} \text{ cm}^{-2}$  at the beam  $1 \mu\text{A/cm}^2$  (Fig.3 curves 4) have caused the shift of onset  $T_c$  to 120 K and significant drop of the resistivity at 100 K  $< 0.1 \mu\Omega$  in 2-YBCO and  $< 1 \mu\Omega$  in 3-GdBCO, which are less than in non-irradiated samples. Since the induced amorphization related to oxygen disordering (transition from superconducting orthorhombic to normal tetragonal phase), it recovered easily (resulting in the initial resistivity below  $\mu\Omega\cdot\text{cm}$  due to a higher oxygen mobility in CuO chains with plenty of oxygen vacancies and large defect instability area [8], when irradiated at the electron current density  $1 \mu\text{A/cm}^2$  to the dose  $10^{15} \text{ cm}^{-2}$ .

Fig.4 *a,b,c* show the resistivity of the non-irradiated and  $\gamma$ -irradiated samples at 77 K.

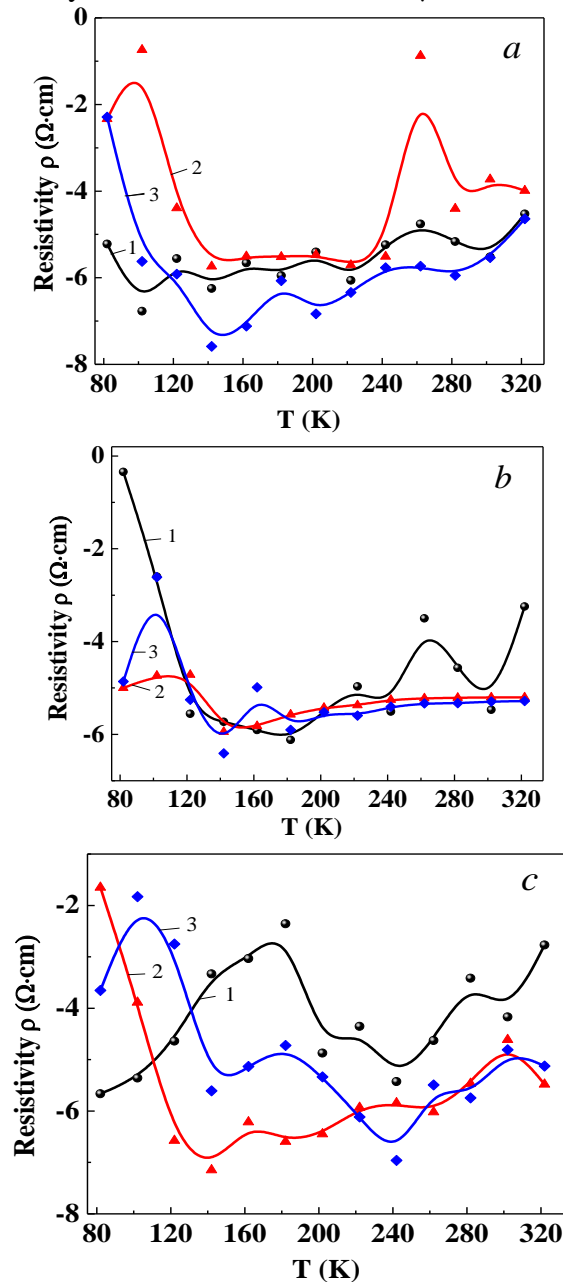


Fig.4. Resistivity of the samples irradiated with  $^{60}\text{Co}$   $\gamma$ - in liquid nitrogen: (a) -1-YBaCuO, (b) -2-YBaCuO, (c) -3-GdBaCuO: 1 – reference, 2 –  $5 \cdot 10^5 \text{ R}$ ; 3– additional dose  $5 \cdot 10^5 \text{ R}$  (total  $10^6 \text{ R}$ )



Comparison of fig. 3 and 4 reveals the common resistivity peak at 100 K growing after the both kinds of irradiations and annealed  $< 200$  K, which can be attributed to paramagnetic defects in Cu-O-chains. This result agrees well with our previous data on  $\gamma$ -irradiations at 77 K [15,21] and other's at 4–77 K caused by positron generation and reviewed in [8]. Unexpected difference in behavior just above  $T_c$  between 1-YBaCuO and 2-YBaCuO. Difference in peak at 260 K for both YBCO and at 280 K for GdBCO can be attributed to defects in double CuO<sub>2</sub> planes, which are spaced differently by non-magnetic Y<sup>3+</sup> and magnetic Gd<sup>3+</sup> [22]. Recent molecular dynamics simulations of radiation damage in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> demonstrated, that during the cascade of fast neutron induced defects, the interstitial sites between CuO chains (fig.1b) are populated with oxygen interstitials [23].

### **XRD structure and phase analyses.**

At first one should emphasize, that XRD identifies the crystal structure and estimates its volume amount in % only for crystalline phases, while amorphous phases are seen as a broad scattering band at low angles. Non-irradiated samples 1-YBaCuO and 2-YBaCuO contain Y<sub>2</sub>O<sub>3</sub> and CuO and 3-GdBaCuO contains Gd<sub>2</sub>O<sub>3</sub> interface nanophases. It means, that the pinning centers in the non-irradiated tapes are oxide nanophases at the interfaces of HTSC. After electron irradiation to  $10^{15}$  cm<sup>-2</sup> there appeared 22% of a new YCuO<sub>2</sub> nanophase in 1-YBaCuO; the superconducting YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> phase increased up 47% at the expense of Y<sub>2</sub>O<sub>3</sub> and CuO phases decrease, the ration of Gd<sub>2</sub>O<sub>3</sub> to GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> increased, i.e. the irradiation induced recrystallization of the interfaces.

### **Conclusions and Perspectives.**

As irradiation with 5 MeV electrons and 1.17–1.33 MeV gamma flux do not produce long living radio-nuclides, it is affordable for industrial technology of radiation treatment of long cable by rewinding across the flux. Thus effect of nuclear irradiation does not always damages structure and degrades functions of materials. There exist particular ranges of energy/intensity/dose/temperature where structure modification may result in improving the properties and even find new functions for old materials.

**Acknowledgements.** The research is supported in parts by Program of fundamental and applied researches for Institute of nuclear physics by President Decree No 4526 of 21.11.2019 and collaboration with CAT and JINR. The authors appreciate Prof. S.I. Tyutyunnikov and M.S. Novikov for providing the industrial HTSC coated tapes and closed interest in the researches.

## **2. References**

1. Bednorz J.G. and Müller K.A. Possible high  $T_c$  superconductivity in the Ba-La-Cu-O system // *Zeitschrift für Physik B Condensed Matter*, 1986, 64 (2), pp. 189–193.
2. Wu M.K., Ashburn J.R., Torng C.J., Hor P.H., Meng R.L., Gao L., Huang Z.J., Wang Y.Q., and Chu C W, Superconductivity at 93 K in a new mixed-phase Y-Ba-Cu-O compound system at ambient pressure // *Physical Review Letters*, 1987, 58 (9), pp. 908–910.
3. Vilém Bartůňek., Jose Luis Pérez-Díaz., Tomáš Hlášek., Ladislav Viererbl., Hana Assmann Vratislavská. Influence of neutron and gamma radiation on YBCO and GdBCO/Ag superconducting bulks // *Ceramics International* 2020, 46, pp. 15400–15407. <https://doi.org/10.1016/j.ceramint.2020.03.085>.
4. Jia Y., LeRoux M., Miller D.J., Wen J.G., Kwok W.-K., Welp U., Rupich M.W., Sathiyamurthy X. Li, S., Flesher S., Malozemoff A.P., Kayani A., Ayala-Valenzuela O.,

- Civale L. Doubling the critical current density of high temperature superconducting coated conductors through proton irradiation // *Appl. Phys. Lett.* 2013, 103, 122601. <https://doi.org/10.1063/1.4821440>.
5. Rupich M.W., Sathyamurthy S., Fleshler S., Solovyov Q.Li,V., Ozaki T., Welp U., Kwok W.-K., LeRoux M., Miller D.J., Kihlstrom K., Civale L., Eley S., Kayani A. Engineered pinning landscapes for enhanced 2G coil wire // *IEEE Trans. Appl. Supercond.* 2016, 26 6601904. <https://doi.org/10.1109/TASC.2016.2542270>.
  6. Troitskii A.V., Demikhov T.E., Antonova L.K., Didik A.Y., Mikhailova G.N. Radiation effects in high-temperature composite superconductors // *J. Surf. Invest. X-ray, Synchrotron Neutron Tech*, 2016, 10, 381–392. <https://doi.org/10.1134/S1027451016020397>.
  7. Umezawa A., Crabtree G.W., Liu J.Z., Weber H.W., Kwok W.K., Nunez L.H., Moran T.J., Sowers C.H., and Claus H. Enhanced critical magnetization currents due to fast neutron irradiation in single-crystal  $\text{YBa}_2\text{Cu}_3\text{O}_{7-b}$  // *Phys. Rev. B: Condens. Matter*, 1987, 36, 7151–7154. doi: <https://doi.org/10.1103/PhysRevB.36.7151>.
  8. Quere Y. Radiation effects in old and new superconductors // *Nucl. Instrum. Meth. B*, 1988, Volume 33, Issues 1–4, pp. 906–912. [https://doi.org/10.1016/0168-583X\(88\)90708-2](https://doi.org/10.1016/0168-583X(88)90708-2).
  9. Jorgensen J.D., Beno M.A., Hinks D.G., Soderholm L., Volin K.J., Hitterman R.L., Grace J.D., Schuller Ivan K., Segre C.U., Zhang K., and Kleefisch M.S. Oxygen ordering and the orthorhombic-to-tetragonal phase transition in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  // *Phys. Rev. B*, 1987, 36, 3608 DOI: <https://doi.org/10.1103/PhysRevB.36.3608>.
  10. Heald S.M., Tranquada J.M., Moodenbaugh A.R., and Xu Youwen Orientation-dependent x-ray-absorption near-edge studies of high- $T_c$  superconductors // *Phys. Rev. B*, 1988, 38, 761. doi:<https://doi.org/10.1103/PhysRevB.38.761>.
  11. Cava R.J., Hewat A.W., Hewat E.A., Batlogg B., Marezio M., Marezio K.M., Rabe K.M., Krajewski J.J., Peck Jr. W.F., and Rupp Jr. L.W. Structural anomalies, oxygen ordering and superconductivity in oxygen deficient  $\text{Ba}_2\text{YCu}_3\text{O}_x$  // *Physica C*, 1990, 165, p.419–433.
  12. Wai-Kwong Kwok., Ulrich Welp., Andreas Glatz., Alexei E Koshelev., Karen J Kihlstrom., George W Crabtree. Vortices in high-performance high-temperature superconductors // *Rep. Prog. Phys*, 2016, 79 (11). 116501 DOI: 10.1088/0034-4885/79/11/116501.
  13. Shanghai Superconductors. Available online: <http://www.shsctec.com/en/introduce/89> (accessed on 10 June 2020).
  14. Tsotsopoulou E., Dy'sko A., Hong Q., Elwakeel A., Elshiekh M., Yuan W., Booth C. and Tzelepis D. Modelling and fault current characterization of superconducting cable with high temperature superconducting windings and copper stabilizer layer // *Energies* 2020, 13(24), 6646. doi:10.3390/en13246646.
  15. Mikhailova G.N., Voronov V.V., Troitsky A.V., Didyk A.Yu., Demikhov T.E., Suvorova E.I. Processing method for high temperature superconductor. 2013. Patent RF 2477900.
  16. Novikov M.S., Keilin V. E., Novikov S. I. Preparation and experimental investigation of heavy-current transposed HTS Conductors // *IEEE Trans. Appl. Supercond*, 2013, Vol. 23, N3, 4 pp. DOI:10.1109/TASC.2013.2237733.
  17. Balatsky A.V., Vekhter I., and Zhu J.X. Impurity-induced states in conventional and unconventional superconductors // *Rev. Mod. Phys*, 2006, 78, pp. 373–433.
  18. Molodyk A., Samoilenov S., Markelov A., Degtyarenko P., Lee S., Petrykin V., Gaifullin M., Mankevich A., Vavilov A., Sorbom B., Cheng J., Garberg S., Kesler L., Hartwig Z., Gavrilkin S., Tsvetkov A., Okada T., Awaji S., Abraimov D., Francis A.,

- Bradford G., Larbalestier D., Senatore C., Bonura M., Pantoja A.E., Wimbush S.C., Strickland N.M. & Vasiliev A. Development and large volume production of extremely high current density  $\text{YBa}_2\text{Cu}_3\text{O}_7$  superconducting wires for fusion // *Scientific Reports* 2021. 11. 2084. (1–11pp). <https://doi.org/10.1038/s41598-021-81559-z>.
19. Chepikov V., Mineev N., Degtyarenko P., Lee S., Petrykin V., Ovcharov A., Vasiliev A., Kaul A., Amelichev V., Kamenev A., Molodyk A. and Samoilnikov S. Introduction of  $\text{BaSnO}_3$  and  $\text{BaZrO}_3$  artificial pinning centres into 2G HTS wires based on PLD- $\text{GdBCO}$  films. Phase I of the industrial R&D programme at SuperOx // *Superconductor Science and Technology*, 2017, Vol.30, (12 pp). doi 10.1088/1361-6668/aa9412.
  20. Hylton T.L., Beasley M.R. Flux-pinning mechanisms in thin films of  $\text{YBa}_2\text{Cu}_3\text{O}_x$  // *Phys. Rev. B Rapid Commun*, 1990, 41, 11669-72 doi:<https://doi.org/10.1103/PhysRevB.41.11669>.
  21. [21] Polyak O.Yu., Ibragimova E.M. The effect of gamma-irradiation on  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  ceramics and monocrystals in the superconducting state // *Phys. Stat. Sol. A*. 1989, 122, k45-k50.
  22. Balatsky A.V., Vekhter I. and Zhu J.X. Impurity-induced states in conventional and unconventional superconductors // *Rev. Mod. Phys*, 2006, 78, pp. 373–433.
  23. Gray R.L., Rushton M.J.D. and Murphy S.T. Molecular dynamics simulations of radiation damage in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  // *Supercond. Sci. Technol*, 2022, Vol. 35, N3, 035010. Doi:10.1088/1361-6668/ac47dc.