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PROSPECTS OF UTILIZATION OF HTSC HAVING HIGH CRITICAL CURRENTS DUE TO NUCLEAR FISSION FRAGMENT TRACKS

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Перспективы использования ВТСП с высокими критическими токами за счет треков осколков деления ядер

Один из наиболее эффективных методов увеличения критического тока связан с образованием сравнительно однородно распределенных в объеме ВТСП треков осколков деления ядер. Деления могут быть вызваны либо за счет облучения частицами высоких энергий в случае ядер Hg, Tl, Pb, Bi, входящих в состав ВТСП, либо за счет облучения нейтронами или гамма-квантами в случае ядер U, введенного в сверхпроводник. Анализируются проблемы оптимального флюенса, радиоактивности и стоимости облучения, а также рассматриваются перспективы использования модифицированных таким образом ВТСП материалов для создания сверхпроводящих магнитных систем.

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Prospects of Utilization of HTSC Having High Critical Currents due to Nuclear Fission Fragment Tracks

One of the most effective methods of increasing a critical current is due to the production of nuclear fission fragment tracks almost uniformly distributed inside HTSC. These fissions can be caused either by high energy particle irradiation for Hg, Tl, Pb, Bi, which belong intrinsically to HTSC, or by neutron/gamma irradiation for doped U nuclei. The problems of optimum radiation fluences, radioactivity and cost are analysed in this paper. The prospects of utilization of such modified HTSC materials for superconducting magnetic systems are also considered.

The investigation has been performed at the Laboratory of High Energies, JINR.

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1. INTRODUCTION

The value of critical current density in HTSC depends on the availability of defects having suitable sizes, properties and orientation, which act as fluxoid pinning centers. One of the most effective types of defects are swift heavy ion tracks, produced by particles, having a linear energy loss of $dE/dx > (5 \div 10)$ keV/nm (see, for example, reviews [1,2]). The tracks can be created not only by accelerator beam irradiation but also due to arising of nuclear fission fragments inside HTSC as a result of external bombardment by suitable particles. In the latter case, the lengths of effective track parts are $\approx(4 \div 7)\mu$m and the transverse dimensions of their amorphous nonsuperconducting cores $\approx(2 \div 8)$ nm.

In principle, HTSC, having $T_c \approx (85 \div 130)$K, can be utilized over a wide range of temperatures $4.2\ K \leq T < T_c$. But the cost of work at 4.2K : 27K : 50K : (62÷77) K corresponds to 100% : 19% : 7% : 5%. So, from the economical point of view and convenience, it is worth-while to use liquid nitrogen temperatures. Unfortunately, the intervals of the magnetic field, in which the values of $J_c(B,T)$ are acceptable, as a rule become narrow or even disappear over this range of temperatures.

The methods of appreciable increasing the values of HTSC $J_c(B,T)$ due to irradiations are considered in this paper.

2. OPTIMUM FLUENCES

First of all, it seems useful to collect many methods in Table 1, where one can see the probabilities of nuclear fissions (σ_f-cross sections, 1 barn$=10^{-24}$ cm^{-2}) and the values of optimum fluences (F is the full flux per cm^2).

The fluence is, by definition, optimum when the ratios $R_J(F) = J_C(B,T,F)/J_C(B,T,0)$ or $R_B(F) = B_{trap}(T,F)/B_{trap}(T,0)$ reach maximum significances (here B_{trap} is a maximum magnetic field which can be trapped in an HTSC object, e.g. a disk).

Table 1. The methods of increasing J_c in HTSC by irradiations.

Method`s legend	Bombarding particles, optimum fluences, cross sections
A	**Without fission of nuclei**
A1	electrons; neutrons ($E_n \geq 0.1$ MeV); light ions ($dE/dx < 2 \div 5$ keV/nm)
A2	heavy ions ($dE/dx > 5 \div 10$ keV/nm); $F_{h.i.}^{optim} = 10^{11} \div 10^{12}$ cm^{-2}
B	**With fission of U nuclei doped into HTSC**
B1	thermal n→U-235, $\sigma_f \approx 582$ barns, $F_{t.n}^{optim} \approx (1 \div 12) \cdot 10^{16}$ cm^{-2}
B2	fast n ($E_n \geq 1.4$ MeV) →U-238, $\sigma_f \approx 0.6$ barns, $F_{f.n}^{optim} > 10^{18}$ cm^{-2}
B3	gamma – quanta ($E_\gamma \geq 15$ MeV) → U-238, $\sigma_f^{max} \approx 0.15$ barns
C	**With fission of heavy nuclei which belong intrinsically to HTSC: Hg, Tl, Pb, Bi**
C1	p, d (E=200-1000 MeV) →A_i; $\sigma_f = 0.1 \div 0.15$ barns, $F_{p,d}^{optim} \approx 10^{17}$ cm^{-2}

Obviously, Fleischer et al [3] first measured $R_J(F_{t.n})$ for YBCO polycrystalline, doped with a natural U mixture containing 1/139 of U-235. Knowing the later results, one can approximately restore their full curves $R_J(F_{t.n})$ and find maximum values and $F_{t.n}^{optim}$ (see Table 2). The authors found that U, distributed nonhomogeneously in the YBCO matrix, was contained in very small particles.[1] It is important to note that an optimum fluence and rather high R_J^{optim} values corresponding to the smallest average fission density $<n_f^{optim}> \approx 10^{14}$ cm^{-3} were observed in case of the lowest U content. These facts can be explained by a more homogeneous and hence more effective space distribution of fission fragment tracks than in case of a higher U content (compare their results for 0.07 and 0.18w. %U).

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[1] Later, Sawh et al [4] showed that uranium was actually contained in $U_2Y_3Ba_5O_{15}$ compound particles in YBCO without Pt.

Table 2. Optimum parameters for (YBCO + U) irradiated with thermal neutrons (method B1).

Total U content $C_{235}+C_{238}$ (w.%)	U-235 content C_{235} (w.%)	Optimum thermal neutron $F_{t.n}^{optim}$ (10^{16}cm^{-2})	Optimum average fission $<n_f^{optim}>$ (10^{14} cm^{-3})	$R_J = \frac{J_c^M(F_{t.n}^{optim})}{J_c^M(0)}$ at B=1T and T'=63K	T''=77K	$R_B = \frac{B_{trap}(F_{t.n}^{optim})}{B_{trap}(0)}$ at T''=77K
			Polycrystalline YBCO + U (work [3])			
0.07	0.0005	≈200	≈1	≈25$^{b)}$	≈13$^{c)}$	–
0.18	0.0013	≈230	≈2.5	≈18	≈8	–
			Melt-textured YBCO+0.5w.%Pt + U (works[5-7])			
0.15	0.15 K_{235}a)	≈12	≈15 K_{235}	–	–	5.1
0.3	0.3 K_{235}	≈10	≈25 K_{235}	–	≈10	4.5$^{d)}$
0.7	0.7 K_{235}	≈7	≈40 K_{235}	–	≈4	3.2

a) $K_{235} = C_{235}/(C_{235}+C_{238})$ b) $J_c^M(1T, 63K; F_{t.n}^{optim}) = 7 \cdot 10^4$ A/cm^2

c) $J_c^M(1T, 77K, F_{t.n}^{optim}) = 1.4 \cdot 10^4$ A/cm^2

d) $B_{trap}^{max}(62K) = 8.1T$; $B_{trap}^{max}(77K) = 3.2T$ for 4 disks of YBCO+0.3w.%U (∅2 cm; h=4·0.8 cm).

One can find the same tendency using the results of the other group of authors [4-6]. They found that it was necessary to add 0.5w.%Pt to make single grain structure YBCO disks (∅2 cm, 0.8 cm in thickness) containing up to 0.8w.%U. Then, U was included in ≈0.3μm $(U_{0.6}Pt_{0.4})YBa_2O_6$ particles. The average distances between these particles depend on w.%U (e.g., they were equal to 3.9 μm for 0.3w.%U and 2.9 μm for 0.6w.%U etc.). Though the authors did not say anything about a relative content of U-235 (K_{235} in our Table 2), one could assume $0.1 \leq K_{235} \leq 0.98$ and evaluate their $<n_f>$ (see Table 2). The found tendency can be seen even more clearly from the experimental results on J_c

increasing after the p(800 MeV) bombardment of Bi-based [8] and Hg-based HTSC [9]. Here, the tracks of Bi and Hg nuclear fission fragments were distributed practically homogeneously resulting in high R_J ratios at modest $n_f^{optimr} \leq 10^{14}\,\mathrm{cm}^{-3}$. Note that the content of elements with fissionable nuclei was ≈44 w.%(Bi, Pb) and ≈27w.%Hg in these cases. The last numbers can explain why the optimum fluences are not very high ($<2 \cdot 10^{17}\,\mathrm{cm}^{-2}$) at small $\sigma(p,f) \leq 0.15$ barns.

3. RADIOACTIVITY PROBLEM

Only the bombardment of HTSC with heavy ions, which energy is lower than the Coulomb barrier, does not result in radioactivity (RA). All other methods of increasing J_c with irradiation lead to the production of isotopes, which mainly emit gamma-quanta from HTSC, fission fragments and matrix over an energy range of $\approx 0.1 \div 2.5$ MeV. An initial radioactivity after irradiation depends on the energy spectrum and type of particles, as well as on element target composition, and it is proportional to fluence. Moreover, all isotopes have different half-lives ($T_{1/2}$). So, in any specific case the full curve of RA decreasing must be calculated (if possible) or measured for some months until a few long-lived isotopes, which make main contributions, remain. One can more or less easily calculate the process for the rest of the time. Taking it into account, let us consider several typical examples[2].

Example 1: thermal n → YBCO + U-235 (method B1)

In this case the radioactivity is $15 \div 25$ % of ^{133}Ba ($T_{1/2} = 10.5$ years, $\sigma_a = 7$ barns, the contribution at 10^{16} t.n/cm^2 is ≈0.5 kBq/g, 1 kBq=10^3decays/s) and the rest of fission fragments in several months of waiting when short-lived isotopes vanish. If the contents of U-235 is $0.1 \div 0.8$ w.% and hence $F_{t.n}^{optim} \cong (1 \div 12) \cdot 10^{16}\,\mathrm{cm}^{-2}$, the full RA level is ≤40 kBq/g. This figure does not contradict the level measured after waiting for 6 months: 7.4 kBq/g for YBCO+0.5w.%Pt + 0.3w.%U and $F_{t.n} = 3 \cdot 10^{16}\,\mathrm{cm}^{-2}$ [6]. Note that it is a small enough RA level: the dose power is not practically different from the natural one at R≈30 cm from a 1gram sample.

Example 2: thermal n→ (Bi2223 + U-235)/Ag (method B1).

As a rule, the multifilamentary composite tape, made by power-in-tube technique, contains more than 80w.%Ag. There are 49% of ^{109}Ag in natural Ag. Much quantity of RA ^{110m}Ag isotope ($T_{1/2} = 250$ days) is produced as a result of the reaction ^{109}Ag(n,γ) ^{110m}Ag (with $\sigma_a \approx 4.7$ barns for thermal n). It is easy to evaluate an initial RA level for 1 gram of Bi 2223/Ag tape (the av. density is 9 g/cm^3) using a simple relation: $A \leq 6.6 \cdot 10^3\, R_F$ (kBq/g), where $R_F = F_{t.n}/10^{16}\,\mathrm{cm}^{-2}$. In 1 month after irradiation with fluences $F_{t.n}(E_n < 0.55\ \mathrm{eV}) = 4 \cdot 10^{16}\,\mathrm{cm}^{-2}$, $F_{f.n}(E_n > 0.1\ \mathrm{MeV}) = 5 \cdot 10^{16}\,\mathrm{cm}^{-2}$, the measured radioactivity levels [10] after recalculating for a 1 gram tape sample, containing ≈20w.% of (Bi2223+0.3w.%U), were: 9000 kBq from Ag, 80kBq from Bi2223 and 18 kBq from fission fragments. They turned out to be in the frame of our prediction; a more precise calculation can be made, if a real energy spectrum of thermal neutrons is known.

Example 3: fast n→ (Bi2223 + U-238)/Ag (method B2).

We attempted to decrease the radioactivity of the Ag matrix using fast neutrons. They practically do not produce ^{110m}Ag though it is nesessary to have ≈10^3 fluences of thermal neutrons for the same fissions per cm^3. The irradiation was made in the vicinity of the active zone of the JINR small reactor IBR-30, where $F_{f.n}/F_{t.n} \approx 5000$. As a result, we obtained the effect of increasing J_c at $B \geq 0.5$T [11] and approximately a 5 times lower radioactivity level from Ag for $F_{f.n}(E_n \geq 1.4\ \mathrm{MeV}) \approx 2 \cdot 10^{17}\,\mathrm{cm}^{-2}$ and 0.82w.%U-238.

Example 4: gamma-quanta (Eγ≤24 MeV)→ (Bi2223 + U-238)/Ag (method B3).

We have also been investigating this photofission method ($\sigma(\gamma,f) \leq 0.15$ barns) because the ^{106m}Ag isotope is produced ($T_{1/2} = 8.2$ days) instead of ^{110m}Ag during this irradiation. First experiments show promising results concerning the RA level.

It is important to make two notes at the end of this section. First, it is most probable that the HTSC, modified by irradiation, will be utilized to produce magnetic fields. In this case, only the inside/central section of windings must be radioactive and the

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[2] The radioactivity levels were evaluated by V.V.Golikov. The autor is pleased to thank for his help as well as for fruitful discussions.

other parts of the magnetic system (outer windings, yoke, cryostat) will serve as an effective shield absorbing gamma-quanta. It is not a problem to use also an additional shield, if necessary. Second, such systems must be extremely reliable in order to have any repair only as an exception. As a rule, they must be made by the organization with experienced personnel and special-purpose equipment.

4. COST OF IRRADIATIONS

The general statement is that HTSC modifications using reactors (except method B2) are at least one order of magnitude cheaper than those using accelerators though there are some other factors which should be taken into account in any practical case.

In principle, there are all sources of irradiations for increasing HTSC J_c at JINR: reactors, heavy ion cyclotrons, proton (660 MeV, 1μA) and electron accelerators. In this report we briefly describe only the JINR IBR-2 pulse reactor. For example, using beam line no.3 with a 20×40 cm^2 window and the large-scale sample irradiation facility [12], one can have a time-averaged thermal neutron flux density of $\approx 10^{12} cm^{-2} s^{-1}$ near the moderator surface (and approximately the same figure for fast neutrons). The IBR-2 reactor usually operates ≤ 10 cycles per year, a 2-week duration of each of them. It is possible to say that even the commercial type of irradiations using this reactor (or also the JINR proton accelerator) has been much cheaper than in the West up to now.

5. CONCLUSIONS

5.1 Careful choosing HTSC irradiation makes it possible to enlarge significantly the magnetic field interval, in which the value of J_c becomes acceptable (in particular, at liquid nitrogen temperatures).

5.2 The analysis, made in this report, shows that the more homogeneously the fissionable nuclei are dispersed in HTSC, the smaller number of fissions per volume unit are required for optimum results in increasing J_c.

5.3 The radioactivity problem turns out to be serious enough only in method B1 (Table 1) for HTSC/Ag composites irradiated with thermal neutrons (because of the ^{110m}Ag isotope of $T_{1/2}$=250 days) and maybe in method C for first months after proton irradiation with $E_p > 200$ MeV. There are some possibilities to make the radioactivity problem easy including, for example, γ→(Bi2223 + U-238)/Ag irradiation (method B3). In any case, the RA level decreases by a factor of 2, 4, 8, 16 … and so on after waiting for $T_{1/2}$, 2 $T_{1/2}$, 3 $T_{1/2}$ etc.

5.4 So, there are some reasons to think that increasing of J_c due to the production fission fragment tracks inside HTSC will be successfully utilized despite the radioactivity problem and cost rise in any significant cases.

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