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13. ABSTRACT (Maximum 200 words) <p style="text-align: center;">The aim of this project was to develop significantly improved high temperature superconductor (HTS) via improved pinning. In particular the work focused on the "U/n process" in which U is added to HTS, and then irradiated with thermal neutrons to fission some of the ²³⁵U. This process raised J_c in YBa₂Cu₃O₇ (Y123) to world record levels. The U/n process was studied in Bi₁Sr₂Ca₂Cu₃O₁₀ (BiSCCO 2223). Again, world record J_c was achieved plus record H_{irr} and isotropy. J_c and isotropy were increased by factors of up to 100, and H_{irr} was more than doubled. U/n processing was also developed for Nd123. This is not yet optimized, but to date J_c > 150,000 A/cm² in Nd123, a world record. The increases in J_c achieved are sizable, not marginal (e.g., a factor of 40 in Y123 at T = 77K, B = 0.25T). It was proven, first theoretically and then experimentally, that in the specific HTS Sm123 and Gd123 the U/n process cannot be used for large objects. The success of U/n pinning has been studied and is found to be attributable to multiple in-line defects – a domain of high-energy ion damage only sparsely studied for HTS characterizations.</p>			
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Sincerely,



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Enclosure 3

2.b.4. Statement of the problem studied

The authors had discovered that excellent current-carrying capability of the high temperature superconductor (HTS) $\text{YBa}_2\text{Cu}_3\text{O}_7$ (Y123) resulted from incorporating uranium in the Y123, and then irradiating with thermal neutrons. The exposure to the high-energy ions resulting from nuclear fission produced superior pinning centers in Y123, stabilized the magnetic field, and thereby increased current. This is so-called U/n processing.

The authors proposed to extend the study of U/n processing to (i) determine the optimum U and neutron fluence in Y123; (ii) test to see if the process could be extended to other HTS (Nd123, BiSCCO, Sm123); (iii) see if other HTS characteristics are also improved by the U/n process (irreversibility field, H_{irr} , isotropy, . . .).

2.b.5. Summary of the most important results

Y123: As proposed, the U/n process was optimized in Y123. Enhancement of J_c was studied vs. variables: U doping percent; $^{238}\text{U}/^{235}\text{U}$ ratio; Pt content; and neutron fluence, F_n . The ions resulting from fission have range $\sim 10 \mu\text{m}$ in Y123. A key number, never previously measured, is what portion of this length, δ , is effective in pinning. The result found was

$$\delta = 2.7 \mu\text{m}$$

(On theoretical grounds the authors had earlier speculated that $2 \leq \delta \leq 4 \mu\text{m}$.)

From the value of δ we learn that deposits of U compound in Y123 should preferably be spaced by a distance $S < 5.4 \mu\text{m} = 2\delta$. Knowing S , one can test the chemistry of candidate HTSs to see if the U/n process will be successful in any given HTS.

The optimum amount of added U was found to be $M_U \sim 0.15 - 0.30\%$ by weight. (This achieved the desired S .) For 250 ppm of ^{235}U , the optimum neutron fluence is $F_n \sim 8 \times 10^{16} \text{ n/cm}^2$. We also confirmed that the optimum value of F_n varies as $1/M(\text{U}^{235})$. For these values, J_c in Y123 is close to $300,000 \text{ A/cm}^2$ at 77K, $B = 0.25\text{T}$, or about 40 times the value of J_c without U/n processing.

BiSCCO-2223: The U/n process was successful in BiSCCO. Several reviewers indicated that in BiSCCO, J_c is limited by weak links, not pinning centers, hence U/n processing would not be successful in BiSCCO. They were incorrect. In fact, one result of this entire project was to demonstrate that *a dirth of pinning centers, not weak links or large angle of intersection for crystal planes, is the dominant factor in restricting J_c for present day bulk HTS.* J_c in BiSCCO is increased by factors as high as 100 by U/n processing. At the same time, H_{irr} is increased by > 2 , and anisotropy is decreased by up to 100 times (e.g., from 300 to 3). We completed studies of BiSCCO 2223 for a large range of F_n , temperature, and B (up to ~ 5 Tesla). We found that U enters the BiSCCO matrix. It does not form precipitates as does U in Y123. We varied the amount of U added, and studied 0.15, 0.3, 0.6, 1.0 and 2.0% by weight of U. The materials behaved similarly but high U mass has an advantage. Since deposits of U are of \sim atomic dimensions, we need not put in added ^{238}U to get the deposits close together ($< 5.4 \mu\text{m}$).

As a result, pure ^{235}U can be used. More ^{235}U means lower fluence is required. The presence of Ag, in Ag-BiSCCO tape, results in non-negligible radioactivity. Increasing the amount of ^{235}U from 250 ppm to, for example, 2.5% reduces the resulting Ag radioactivity by a factor of 100. Hence high masses of ^{235}U are desirable. Our collaborator, S.X. Dou, found that even 2% U did not degrade I_c by more than 20%.

Nd123: Making the U/n process work in Nd123 proved very difficult. It has only recently been accomplished, and the resulting publication is now being written.

The problem is that when U is added to Nd123, large deposits ($d \sim 5 \mu\text{m}$) of UBa_3O_6 form. For the maximum chemically acceptable amounts of U, these deposits are separated by large distances ($S \gg 5.4 \mu\text{m}$). We will not recite here all of the failed attempts we made to reduce the size of the deposits, but these included additions of Pt, and two externally fabricated double perovskites, $(\text{U}_{0.6}\text{Pt}_{0.4})\text{YBa}_2\text{O}_6$ and $(\text{U}_{0.6}\text{Pt}_{0.4})\text{NdBa}_2\text{O}_6$.

The solution to the problem was found accidentally. We "started over," adding only U to Nd123. The large deposits of UBa_3O_6 again dominated. However, a few very small deposits were seen. These proved to be compounds of U and Zr. We wanted small deposits. However, we had not added Zr. However, during melt texturing, the Nd123 was supported on Zr rods. Evidently, some of the Zr had migrated up into the Nd123. We then did a series of experiments using ZrO_2 in varying amounts. We found that about 1/2 of 1% Zr gave spacing $S < 5.4 \mu\text{m}$, and was therefore suitable for U/n-Nd123 studies. A first "quick look" at U/n-Nd123 is complete. It shows trapped field enhanced by a factor of over 6. This corresponds to $J_c > 150,000 \text{ A/cm}^2$, a world record. The final results will exceed this due to (a) better Nd123; (b) a better determination of peak fluence; and (c) we will correct the temperature profile for melt texturing, to allow for changes in melting temperature due to the presence of U and Zr. All of this is now easily done.

A bonus of the U/n process was the discovery that Zr alone, in Nd123, strongly "refines" the ever-present $\text{Nd}_4\text{Ba}_2\text{Cu}_2\text{O}_{10}$ deposits in Nd123, so that these deposits also act as pinning centers. We therefore now have a "three stage" pinning phenomenon. (1.) Zr refines Nd422, so that Nd422 forms pinning centers. (2.) U,Zr deposits form. These also act as pinning centers. (3.) Thermal neutron irradiation fissions the ^{235}U isotope. The damage done by the fission ion acts as strong pinning centers.

Sm123 and Gd123: A theoretical study was done on thermal neutrons in Sm and Gd. We found, and published, that in these materials the neutrons were absorbed so quickly, by (n,γ) interactions, that not enough remained to satisfy the need to fission ^{235}U . Subsequently, an experiment was run on U + Sm123, and we confirmed that, upon neutron irradiation, only an outer skin of Sm123 was improved.

Summary and New Work

1. Work Completed. All of the proposed work was completed. The U/n process works well in Y123, BiSCCO 2223, and Nd123. Wherever the chemistry of the HTS permits the U/n process to be used, and the (n,γ) cross sections of the HTS do not swallow the neutrons, the U/n process results in world record J_c , H_{irr} , and isotropy.

2. MILD Pinning. The above record J_c , H_{irr} , isotropy, etc. prompted us to study the damage in the U/n process, which is so effective in pinning. A proposal reporting this study has been submitted to DOE and NSF, and provided to ARO for perusal. A key variable is identified, namely the energy loss per Angstrom, S_e , by electron collisions of an energetic ion. Evidence shows that very high J_c results for $0.8 < S_e < 1.8$ keV/Å. The great majority of the work done studying HTS properties after ion damage has, however, been done in the interval $2.5 < S_e < 3.5$ keV/Å. We showed that in the latter (studied) interval, attempts to increase J_c , to pin fields $B > 7.2T$, actually reduce J_c to zero. Somewhere in the region $0.8 < S_e < 2.5$ keV/Å an optimum exists. In this region multiple in-line defects (MILD) are formed. MILD pinning centers are responsible for present world record J_c , H_{irr} and isotropy found in fission ion irradiation. We estimate that MILD pinning will increase J_c in textured, thick film, or crystalline HTS to levels presently found only in thin film HTS. We are in hot pursuit of this possibility.

3. Chemical Pinning. We have noted the unwillingness of some industries to produce products including U. Motivated by this we have found a substitute for U in the *chemical* pinning centers $(U_{0.6}Pt_{0.4})YBa_2O_6$. W has been used to substitute for U. It forms $(W_{0.5}Pt_{0.5})YBa_2O_6$. The effect of (W,Pt) pinning centers on J_c is presently being tested. Mo has also been observed to form small pinning centers. These are under study.

4. Collaborators. This work has been done in collaboration with Prof. S.X. Dou, Univ. of Wollongong, NSW, Australia, who supplied the Ag-BiSCCO tape made of powders we mixed, and with Prof. Harald Weber, Atominstitut, U. of Austria, Vienna who provided VSM studies of Y123 and BiSCCO. Prof. Masato Murakami of SRL, ISTEK, Tokyo transferred to us the technology for melt-texturing Nd123 and Sm123. We transferred technology to American Superconductor, Westborough, MA. (on U/n BiSCCO), M.D. Anderson Cancer Center, Houston, TX. (on the separation of cancer cells by magnetic means), and Aquafine, Corp., Brunswick, GA. (on the clarification of kaolin clay by laminar flow magnetic separation).

2.b.6. List of publications

- Damian G. Marinaro, Shi X. Dou, Josip Horvat, John Boldeman, Roy Weinstein and Ravi Sawh, "Effect of Uranium Doping and Thermal Neutron Irradiation on the Flux-Pinning of Silver-Clad Bi-Sr-Ca-Cu-O Tapes," IEEE Transactions on Applied Superconductivity, Vol. 11 (March 2001), 3896.
- R. Weinstein, R.-P. Sawh, J. Liu, D. Parks, Y. Ren, V. Obot, and C. Foster, "Threshold for creation of ionization pinning centers in YBCO by heavy ions," presented at the 13th International Symposium on Superconductivity (ISS 2000), Tokyo, Japan, October 15, 2000. *Physica C* 357 (2001) 743.
- R. Weinstein, Y. Ren, R. Sawh, A. Gandini, W. Hennig, M. Murakami, T. Mochida, N. Chikumoto, N. Sakai, G. Krabbes, W. Bieger, D. Milliken, S.X. Dou, S. Tönies, M. Eisterer and H.W. Weber, "Properties of HTS for Successful U/n Processing," *Physica C* 341 (2000) 1415.

- W. Hennig, D. Parks, R.-P. Sawh, and R. Weinstein, "Enhanced Levitation Force Using $\text{YBa}_2\text{Cu}_3\text{O}_y$ Trapped Field Magnets," *Physica C* **341** (2000) 2613.
- A. Gandini, R. Weinstein, Y.R. Ren, R.P. Sawh, D. Parks, Y.C. Guo, B. Zeimet, S.X. Dou, S. Tönies, C. Klein, and H.W. Weber, "Critical Current Enhancement in $(\text{Bi,Pb})_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ Tapes Via Isotropic Quasi-Columnar Defects, induced by Uranium Fission Products," *Physica C* **341** (2000) 1453.
- R.P. Sawh, R. Weinstein, Y. Ren, V. Obot and H.W. Weber, "Uranium Fission Fragment Pinning Centers in Melt-Textured YBCO," *Physica C* **341** (2000) 2441.
- M. Eisterer, S. Tönies, H.W. Weber, R. Weinstein, R. Sawh, Y. Ren, "High Critical Currents due to Fission Tracks in $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$," *Physica C* **341** (2000) 1439.
- S. Tönies, H.W. Weber, D. Milliken, Y.C. Guo, S.X. Dou, A. Gandini, R. Sawh, Y. Ren, R. Weinstein, "Influence of neutron irradiation on the superconducting properties of BiSCCO-tapes containing different amounts of uranium," *Physica C* **341** (2000) 1427.
- D. Marinaro, S.X. Dou, J. Horvat, Y.C. Guo, J. Boldeman, A. Gandini, R. Weinstein, R. Sawh, Y. Ren, "The Effects of Uranium Doping and Thermal Neutron Irradiation on the Pinning Properties of Ag/Bi-2223 Tape," *Physica C* **341** (2000) 1119.
- W. Hennig, D. Parks, R. Weinstein, R.-P. Sawh, and Y. Ren, "Stable Magnetic Levitation with Adjustable Ratio of Levitation Force to Restoring Force using Rings of Zero Field Cooled $\text{YBa}_2\text{Cu}_3\text{O}_y$ Samples," *Supercond. Sci. Tech.* **13** (2000) 1447.
- W. Hennig, R. Weinstein, D. Parks, R-P Sawh, Y. Ren, "Enhanced Levitation Force Using $\text{YBa}_2\text{Cu}_3\text{O}_y$ Trapped Field Magnets," 2nd PASREG Conference, Morioka, Japan, Oct. 19-22, 1999, *Supercond. Sci. and Tech.* **13** (2000) 861-865.
- R. Weinstein, Invited Paper, "An Overview of U/n Processing," Proc. of 12th International Symposium of Superconductivity (ISS-99) Morioka, Japan, Oct. 17-19, 1999, *Advances in Superconductivity XII*, Springer-Verlag, (2000) 521-526.
- S.X. Dou, Y.C. Guo, D. Marinaro, J.W. Boldeman, J. Horvat, P. Yao, R. Weinstein, A. Gandini, R. Sawh, and Y. Ren, "Effect of U-doping level on flux pinning and radioactivity in silver-clad Bi-2223 tapes using thermal neutron irradiation," *Advances in Cryogenic Engineering*, **46**, pg 761 (2000). [ICMC-99 conference, Montreal, July 12-16, 1999].
- R. Weinstein, Selected Paper, "Increase in J_c and Irreversibility Field, and Decrease in Anisotropy and Lowered Radioactivity in $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ Irradiated by ^{235}U Fission Fragments," 9th International Workshop on Critical Currents (IWCC-99) University of Wisconsin, Madison, WI, July 7-10, 1999.
- R. Weinstein, Invited Talk, Workshop on Bulk High-Temperature Superconductors and Their Applications, Argonne National Lab, May 17-18, 1999.

- G.W. Schulz, C. Klein, H.W. Weber, S. Moss, R. Zeng, S.X. Dou, R. Sawh, Y. Ren, and R. Weinstein, "Enhancement of transport critical current densities in $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ tapes by fission tracks," *Appl Phys Lett* **73**, (28 Dec 98) p. 3935.
- R. P. Sawh, Yanru Ren, Roy Weinstein, Wolfgang Hennig and Takayuki Nemoto, "Uranium Chemistry and Pinning Centers in HTS," *Physica C*, **305**, (1998) 159.
- R. Weinstein, R. Sawh, Y. Ren, W. Hennig, T. Nemoto and H. Weber, "Broken Columnar Pinning Centers from U Fission and the Achievement of $J_c \sim 3 \times 10^5 \text{A/cm}^2$ at 77K in Textured Y123," *Proc. of CIMTEC Conf., Florence, Italy (June 98). Advances in Science and Technology: Science and Engineering of HTS Superconductivity*, **23** (1999) 83.
- W. Hennig, D. Parks, R. Weinstein and R.-P. Sawh, "Enhanced levitation forces with field cooled $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$," *Appl Phys Lett*, **72**, (1998) 3059.
- Roy Weinstein, Ravi Sawh, Yanru Ren, Michael Eisterer, Harald W. Weber, "The role of uranium chemistry and uranium fission in obtaining ultra-high J_c in textured Y123." *Supercond. Sci. Technol*, **11** (1998) 959.
- M. Eisterer, S. Tönies, W. Novak, H.W. Weber, R. Weinstein and R. Sawh, "Improvement of critical current densities by fission tracks in U-doped high temperature superconductors." *Supercond. Sci. Technol*, **11** (1998) 1001.

2.b.7. List of Participating Scientific Personnel

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1. Prof. Roy Weinstein, UH, Physics
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3. Assoc. Prof. Victor Obot, TSU, Mathematics⁽¹⁾

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3. Ravi Sawh, B.S.
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(2) Ph.D. received Aug. 2001

(3) M.S. received May 2000

(4) Ph.D. received Sept. 1999

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10. Erik Westfall
11. Joe Longoria
12. Rosalie Trevino
13. Mike Vasilescu
14. Digant Jariwala
15. Adrian Johnson
16. Barbara Rodriguez

2.b.8. Report of Inventions

- Laminar Flow Magnetic Separation
- High Yield Magnetic Red Blood Cell Separation
- Magnetic Malaria Diagnosis
- Multiple In-Line Defects as Pinning Centers