Effects of Swift-Particle Irradiations on Critical Current Density in CaKFe$_4$As$_4$

A Takahashi$^1$, S Pyon$^1$, S Okayasu$^2$, S Ishida$^3$, A Iyo$^3$, H Eisaki$^3$, M Imai$^1$, H Abe$^4$, T Terashima$^4$ and T Tamegai$^1$

$^1$Department of Applied Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
$^2$Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
$^3$National Institute of Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan
$^4$National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, Ibaraki 305-0047, Japan

E-mail: ayumu-takahashi141@g.ecc.u-tokyo.ac.jp

Abstract. Introduction of columnar defects to superconductors through swift-particle irradiation enhances their critical current density ($J_c$). Iron-based superconductors (IBSs) have been investigated as promising materials for practical applications because of their large $J_c$ at high magnetic fields and temperatures. Recently, another promising IBS CaKFe$_4$As$_4$ (1144-type IBS) was found, and attracts much interest due to its high $J_c$ in the pristine sample. We compare effects of 800 MeV Xe, 3 MeV proton, and 320 MeV Au irradiations on the critical temperature ($T_c$) and $J_c$ of CaKFe$_4$As$_4$ single crystals, and compare them with irradiation effects in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$.

1. Introduction

Introduction of columnar defects to superconductors through swift-particle irradiation enhances their critical current density ($J_c$) [1-4]. It has been demonstrated that the maximum value of $J_c$ and the corresponding dose depend on ion species and its energy [5]. The difference in the optimum dose among ion species may originate from the different diameters and lengths of created defects.

Iron-based superconductors (IBSs) have been investigated as promising materials for practical applications because of their large $J_c$ at high magnetic fields and temperatures. In previous studies, remarkable effects have been demonstrated in IBSs by irradiating heavy ions and protons into Co or K doped BaFe$_2$As$_2$ (Ba-122) single crystals [3, 5]. Recently, another promising IBS CaKFe$_4$As$_4$ (1144-type IBS) was found [6]. Its crystal structure is similar to 122-type IBSs. CaKFe$_4$As$_4$ has a tetragonal structure (P4/mmm), where Ca and K layers stack alternatively along the c-axis [6, 7]. CaKFe$_4$As$_4$ shows similar superconducting properties, such as critical temperature ($T_c$) or upper critical field ($H_{c2}$), to those of optimally K-doped Ba-122-type IBS Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ [8-10]. However, alternate stacking of Ca and K along the c-axis may lead to different physical properties.

Here, we compare effects of 800 MeV Xe, 3 MeV proton, and 320 MeV Au irradiations on the $T_c$ and $J_c$ of CaKFe$_4$As$_4$ single crystals, and compare them with irradiation effects in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. 

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2. Experimental Methods

CaKFe₄As₄ single crystals were synthesized by FeAs self-flux method. Ca granules (99.5%), K ingots (99.5%), and FeAs powder were used as starting materials. FeAs was prepared by sealing stoichiometric amounts of As grains (7N) and Fe powder (99.9%) in an evacuated quartz tube and reacting them at 700 °C for 40 h after heating at 500 °C for 10 h. A mixture with a ratio of Ca : K : FeAs = 1 : 1.2 : 10 was placed in an alumina crucible in an argon-filled glove box. The alumina crucible was sealed in a niobium tube using arc melting method. The niobium tube was sealed in an evacuated quartz tube. The whole assembly was heated for 5 h at 1180 °C after a preliminary heating at 650 °C for 5 h, and cooled to 1050 °C at a rate of 26 °C/h, followed by cooling to 930 °C at a rate of 1.5 °C/h for the crystal growth.

800 MeV Xe and 3 MeV proton irradiations were performed at NIRS-HIMAC, and 320 MeV Au irradiation was performed using the tandem accelerator in JAEA. The irradiation dose is counted by the dose-equivalent magnetic field called “matching field”, where each defect is occupied by single vortex

\[ B_0 = n\Phi_0. \]  

Here, \( n \) is the areal density of defects and \( \Phi_0 \) is a flux quantum.

Magnetization of the crystal was measured by a superconducting quantum interference device (SQUID) magnetometer (MPMS-5XL, Quantum Design). The single crystal was placed in a quartz sample holder and fixed with Apiezon N grease. \( T_c \) was obtained from zero-field cooling (ZFC) and field-cooling (FC) magnetization measurements for field perpendicular to the \( ab \)-plane. \( J_c \) was calculated from the results of the magnetization measurements using extended Bean model.

3. Results and Discussion

3.1. 800 MeV Xe irradiation

![Figure 1](image_url)

**Figure 1.** \( B_0 \) dependences of (a) \( T_c \) and (b) \( J_c \) at 2 K under zero field of 800 MeV Xe-irradiated CaKFe₄As₄ and Ba₀.₆K₀.₄Fe₂As₂.

Figure 1 (a) shows \( B_0 \) dependences of \( T_c \) of 800 MeV Xe-irradiated CaKFe₄As₄ and Ba₀.₆K₀.₄Fe₂As₂. It has been demonstrated that the suppression of \( T_c \) of 800 MeV Xe-irradiated Ba₀.₆K₀.₄Fe₂As₂ is 0.028 K/T, and is smaller than that of samples irradiated with other species [5]. It can be said that this tendency holds in the case of CaKFe₄As₄ since the suppression of \( T_c \) of 800 MeV Xe-irradiated CaKFe₄As₄ is 0.009 K/T, which is even smaller than that of Ba₀.₆K₀.₄Fe₂As₂. Irradiation-resistant feature in CaKFe₄As₄ could be related to the presence of novel planar defects in this material [11]. Figure 1 (b) shows \( B_0 \) dependences of \( J_c \) of 800 MeV Xe-irradiated CaKFe₄As₄ and Ba₀.₆K₀.₄Fe₂As₂ at 2 K under zero field. In the case of 800 MeV Xe irradiation, \( B_0 \) dependences of \( J_c \) in both samples are similar, where \( J_c \) is enhanced up to ~15 MA/cm².
3.2. 3 MeV proton irradiation
Figure 2 (a) shows $B_0$ dependences of $T_c$ of 3 MeV proton-irradiated CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. At least in low-dose region, the suppression of $T_c$ of 3 MeV proton-irradiated CaKFe$_4$As$_4$ is 0.34 K/(1x10$^{16}$ ions/cm$^2$), and is similar to that of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. Figure 2 (b) shows $B_0$ dependences of $J_c$ of 3 MeV proton-irradiated CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ at 2 K at $H$ = 1 kOe. In the case of 3 MeV proton irradiation, $B_0$ dependences of $J_c$ in both samples are also similar, at least in low-dose region. It is expected that higher $J_c$ is obtained from 3 MeV proton-irradiated CaKFe$_4$As$_4$ with higher dose (> 2x10$^{16}$ ions/cm$^2$), similar to the case of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$.

3.3. 320 MeV Au irradiation
Figure 3 (a) shows $B_0$ dependences of $T_c$ of 320 MeV Au-irradiated CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. Suppressions of $T_c$ of both samples irradiated with 320 MeV Au are similar (0.11 K/T). Figure 3 (b) shows $B_0$ dependences of $J_c$ of 320 MeV Au-irradiated CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ at 2 K under zero field. In the case of 320 MeV Au irradiation, $J_c$ in both samples show similar $B_0$ dependences,

Figure 2. Dose dependences of (a) $T_c$ and (b) $J_c$ at 2 K at $H$ = 1 kOe of 3 MeV proton-irradiated CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$.

Figure 3. $B_0$ dependences of (a) $T_c$ and (b) $J_c$ at 2 K under zero field of 320 MeV Au-irradiated CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. The thicknesses of CaKFe$_4$As$_4$ corresponding to $B_0$=2 T, 4 T, 8 T, and 16 T are 9.2 μm, 6.4 μm, 5.5 μm, and 5.4 μm.
CaKFe$_4$As$_4$

Figure 4. (a) Temperature dependence of normalized magnetization ($M$) at 5 Oe and (b) magnetic field dependence of $J_c$ at 2 K of 320 MeV Au-irradiated CaKFe$_4$As$_4$ with the same dose ($B_0 = 4$ T) but with different thicknesses.

where $J_c$ is enhanced with increasing $B_0$ up to $B_0 = 4$ T and almost retains the maximum value up to $B_0 = 16$ T. However, the maximum value of $J_c$ of 320 MeV Au-irradiated CaKFe$_4$As$_4$ is higher than that of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. It is expected that higher $J_c$ close to 30 MA/cm$^2$ could be obtained by introducing splayed columnar defects or coexisting of columnar and point defects, which enhance $J_c$ in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ [12, 13].

We compare the value of $T_c$ and the maximum value of $J_c$ of 320 MeV Au-irradiated CaKFe$_4$As$_4$ with different thicknesses. Figure 4 (a) shows temperature dependence of normalized magnetization ($M$) at 5 Oe of 320 MeV Au-irradiated CaKFe$_4$As$_4$ with the same dose ($B_0 = 4$ T) but with different thicknesses. The value of $T_c$ of thin samples irradiated with 320 MeV Au tends to be lower than that of thick samples. Figure 4 (b) shows magnetic field dependence of $J_c$ at 2 K of 320 MeV Au-irradiated CaKFe$_4$As$_4$ with the same dose ($B_0 = 4$ T) but with different thicknesses. Although the relative error in estimating the thickness of samples becomes larger in thin samples ($t < 7 \mu$m), it can be said from Fig. 4 (b) that the maximum value of $J_c$ of thin CaKFe$_4$As$_4$ samples irradiated with 320 MeV Au tends to be higher than that of thick samples ($t > 12 \mu$m). The projected range of 320 MeV Au in CaKFe$_4$As$_4$ is about 17 \mu m. Thus, in thick CaKFe$_4$As$_4$ samples over 10 \mu m irradiated with 320 MeV Au, defects may not be created through the whole crystal, and $J_c$ in the bottom side of the crystal can be much lower than that in the top side of the crystal. Thickness dependence of $T_c$ suppression can be also explained by the inhomogeneous creation of defects. It should be noted that similar thickness dependences of $T_c$ suppression and $J_c$ distribution due to inhomogeneous creation of defects by low-energy 300 MeV Xe irradiation have been reported for Ba(Fe,Co)$_2$As$_2$ [14].

4. Summary

We compare effects of 800 MeV Xe, 3 MeV proton, and 320 MeV Au irradiations on the $T_c$ and $J_c$ of 1144-type IBS CaKFe$_4$As$_4$ and 122-type IBS Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ single crystals. With respect to the suppression of $T_c$, 3 MeV proton and 320 MeV Au irradiations have similar effects on CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. 800 MeV Xe irradiation suppresses $T_c$ of CaKFe$_4$As$_4$ a little less than that of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. With respect to $B_0$ dependences of $J_c$, irradiated CaKFe$_4$As$_4$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ show similar dependences in all cases. However, in the case of 320 MeV Au irradiation, the maximum value of $J_c$ of CaKFe$_4$As$_4$ is higher than that of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. In addition, the maximum value of $J_c$ of CaKFe$_4$As$_4$ irradiated with 320 MeV Au strongly depends on the thickness of samples and thin samples below 10 \mu m tend to show high $J_c$ over 20 MA/cm$^2$. 

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