# LOW TEMPERATURE NEUTRON IRRADIATION EFFECTS ON TENSILE CHARACTERISTICS OF IRON SINGLE CRYSTALS

KITAJIMA, Kazunori
Research Institute for Applied Mechanics, Kyushu University : Professor

ABE, Hironobu
Research Institute for Applied Mechanics, Kyushu University : Research Associate

TSUKUDA, Noboru
Research Institute for Applied Mechanics, Kyushu University : Research Associate

https://doi.org/10.5109/7173507

出版情報：Reports of Research Institute for Applied Mechanics. 22 (70), pp.209-222, 1975. 九州大学応用力学研究所
バージョン：
権利関係：

Reports of Reseacrh Institute for Applied Mechanics
Vol. XXII, No. 70, 1975

# LOW TEMPERATURE NEUTRON IRRADIATION EFFECTS ON TENSILE CHARACTERISTICS OF IRON SINGLE CRYSTALS

By Kazunori KITAJIMA*, Hironobu ABE**, and Noboru TSUKUDA**

**Abstract**

Measurements were made on yield stress and their recovery characteristics of single crystals of pure iron and iron containing carbon of 10∼50 ppm irradiated in reactor to dose of $6\times10^{16}$ nvt (∼1 MeV) at∼16°K. At low temperatures <90°K, as irradiated specimen showed softening of yield stress, which recovered by annealing near 100°K with activation energy of about 0.26 eV. Appreciable hardening was observed by annealing in the range of 50∼100 °C. Compared with high pure iron, iron containing carbon shows larger hardening at higher temperature range. Mechanisms for softening and hardening were discussed.

## 1. Introduction

So many studies have been made for irradiation hardening of iron and steels since it concerns with the safety of high pressure vessel used in power reactors. However fundamental mechanisms of the irradiation hardening are yet in a stage of controversy. The problems to be solved may be: (1) What is the entity of lattice defects responsible for irradiation hardening, (2) what is the mechanism of the formation of the lattice defects, and (3) how is explained the mechanism of hardening based on interactions of the defects with dislocations.

Low temperature irradiation studies so far developed have succeeded in clarifying some fundamental mechanisms of simplest type of defects, interstitial and vacancy, but much ambiguities remain unsolved not only for the simplest types but also particularly for clusters of defects including impurity atoms [1].

As for the effects of irradiation on mechanical properties of iron, electron irradiation may provide useful tool since it produces rather simplest type defects. Sato and Meshii showed that the interactions of self-interstitials with dislocations result in softening of yield stress at very low temperatures [2)4)5)] and gave a explanation based on their theory of softening in BCC

* Professor, Research Institute for Applied Mechanics, Kyushu University.
** Research Associate, Research Institute for Applied Mechanics, Kyushu University.

metals[3]. similar softening was also found in iron irradiated by neutron at low temperatures by Groh on polycrystals[6] and Kitajima on single crystals[7][8]. Their works include the effects of annealings after irradiation.

On the other hand large effects of impurity elements, paticularly of interstitial impurities on irradiation hardening of BCC metals have been emphasized[9][10][11][12][13]. In these works lattice defects responsible for the hardening are surmised to be some complex defects, *i.e.* irradiation defects combined with interstitial impurity atoms, which are formed at temperature range higher than room temperature, though the entity of the defects is not yet clear.

To resolve the complexities in the problem, more systematic studies are considered to be required at present. This paper intends to provide some fundamental informations for mechanisms of softening and hardening in single crystals of pure iron containing controlled amount of carbon, through measurements of stress-strain characteristics of the specimens after irradiation of neutron at low temperature together with the effects of various post-irradiation annealings on them.

## 2. Experimental Procedure

### 2.1. Specimen

The starting material used in the present investigation was the vacuum-melted re-electrolytic iron supplied by the Japan Iron and Steel Institute. The chemical composition of residual impurities in this material is listed in Table 1. A rod of about 12 mm in diameter was zone-melted by one and 4 passes in atmosphere of wet and dry hydrogen respectively. The ingot was then cold-drawn down to the size of 1 mm in diameter by repeating 3 times cold working and intermediate annealings to obtain uniform structure of grain size of $\sim$0.1 mm. Single crystals were produced by the strain anneal method.
All specimens were then decarburized by annealing in wet hydrogen, and further in high pure hydrogen filtered by palladium foil at 450 °C for several days. By these treatments residual interstitial impurities such as carbon and nitrogen are estimated to be reduced to$<$0.1 ppm. On the other hand, controlled amount of carbon was doped into some of the specimens by annealing them in a flow of hydrogen gas passed through liquid normal heptane. Contents of carbon were estimated by measuring internal friction of monitor plates. Thus the specimens of 10 ppm and$\sim$50 ppm carbon were prepared.

Table 1. Chemical analysis of re-electrolytic pure iron

| C | Si | Mn | P | S | Ni | Cr | Cu |
|---|---|---|---|---|---|---|---|
| .002 | .0015 | .0020 | .0009 | .004 | .0040 | .0012 | .0006 |
| Mo | W | V | Co | Ti | Al | N | O |
| .0002 | .0002 | .0002 | .0098 | .009 | .0005 | .0024 | .0015 |

The single crystals were cut into the length of 25 mm and silver-soldered with end pieces at both ends. The gauge length of the tensile test specimens was 20 mm.

### 2.2. Neutron irradiation and tensile tests

The specimens were irradiated in the low temperature loop of Kyoto University Reactor at 16 °K for 70 hr to the total neutron dose of $3\times10^{16}$ nvt (>1 MeV). After irradiation the specimens were inserted to liquid nitrogen container and brought back to Radio Isotope Laboratory of Kyushu University by air transport.

Tensile tests were carried out on the items: Measurements of stress-strain curves at temperatures of 77°K and 90°K. Measurements for the effects of post irradiation annealings at temperatures up to 200°K on yield stresses at 90°K, and those of post irradiation annealings up to higher temperatures on yield stresses at 200°K. Tests at 77°K and 90°K are made in the bath of liquid nitrogen and oxygen respectively, and those at 200°K in the bath of ethyl alcohol cooled by dry-ice. Annealings of specimen at programed temperatures were performed in the bath of liquid propane for the range of 90°K～200°K, ethyl alcohol for 200～350°K, water for 350～373°K and silicone oil for above 373°K. Isochronal annealing tests were made on same specimen by measuring the yield stresses after successive annealings for 30 minuits increasing temperatures by step of about 20°K. The yield stresses were determined as the flow stress saturated approximately for increments of strain after yield drop if it presents. Residual strains for each test were about 0.3～0.5 %. The test temperature of 90 °K was adopted in order to avoid occurrence of twinning and fracture on one hand and appreciable recovery on the other hand.

## 3. Experimental results

Fig. 1[7] demonstrates the general characteristics of the stress-strain curves of single crystals with 2 mm in diameter tested at 77°K, where the specimens were $ZrH_2$ purified and have the same orientation shown in the figure. Compared with that of the unirradiated specimen, the flow stress of the low temperature irradiated specimen shows hardening at small strain and softening at larger strain. This is sharp contrast to the large hardenings observed in the specimen irradiated at ambient temperature. On the other hand, the specimen irradiated at low temperature and then annealed at 60°C for 30 minutes shows hardening similar to that irradiated at ambient temperature, then fractured by cleavage. This may show that the lattice defects responsible for the hardening are formed at high temperatures. In order to pursue the mechanisms we studied the effects of post-irradiation annealings on the low temperature irradiated specimens.

Fig. 2 shows variations of the magnitude of recovery of softening $\Delta\sigma$.

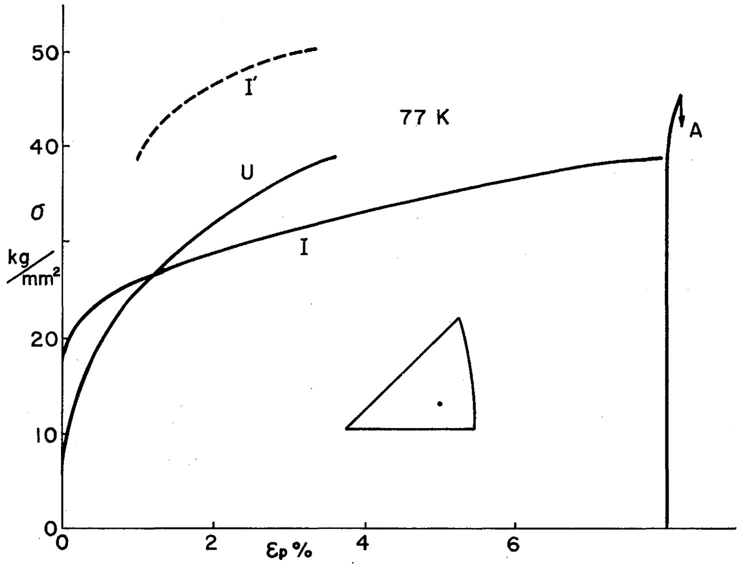


Fig. 1 The stress-strain curves of high purity iron single crystals tested at 77°K. U; unirradiated specimen, I′; ambient temperature irradiated specimen, I; low temperature irradiated specimen, and right hand curve; the same specimen annealed at 340°K for 30 min and retested at 77°K.

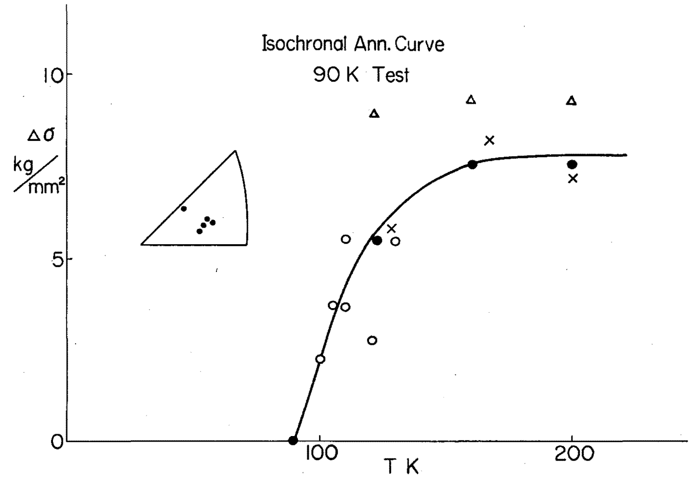


Fig. 2 Isochronal annealing curves in 90∼200°K on the recovery of 90°K yield stress of low temperature irradiated pure iron single crystals.

defined as the increment of yield stress measured at 90°K after isochronal annealings of about 30 min. The remarkable recovery of softening takes place around 100 °K, and is followed by saturation to the value of flow stress in unirradiated specimen near 200°K. Fig. 3 shows similar isochronals of single crystals of pure iron and iron containing 10 ppm carbon with same crystal orientation. Fig. 4 shows the relations between the saturated value of $\Delta\sigma$ and the flow stress of as irradiated specimen $\sigma_Y$ measured at 90°K on some groups of specimens. The group in upper left region of the figure corresponds to the specimen containing 50 ppm carbon. This shows that the softening is suppressed in specimens containing large amount of carbon in contrast to those containing small amount of carbon. In order to obtain further informations about the recovery stage near 100°K, isothermal annealing tests were carried out at the temperatures of 100°K, 105°K and 110°K as shown in Fig. 5. The activatoin energy of the recovery determined from the 3 curves is about 0.26 eV.

On the other hand isochronal annealing curves for the recovery at the temperatures higher than 200°K are shown in Fig. 6 and 7. The main characteristics of the recovery stage are that, stages of hardening near 200 °K and 370°K appear in pure iron, while stage near 200°K is suppressed but large hardening accompanying large upper yield stress and sharp yield drop appear in iron containing large amount of carbon Fig. 6, but the variation is small in iron containing small amount of carbon, Fig. 7. Isothermal

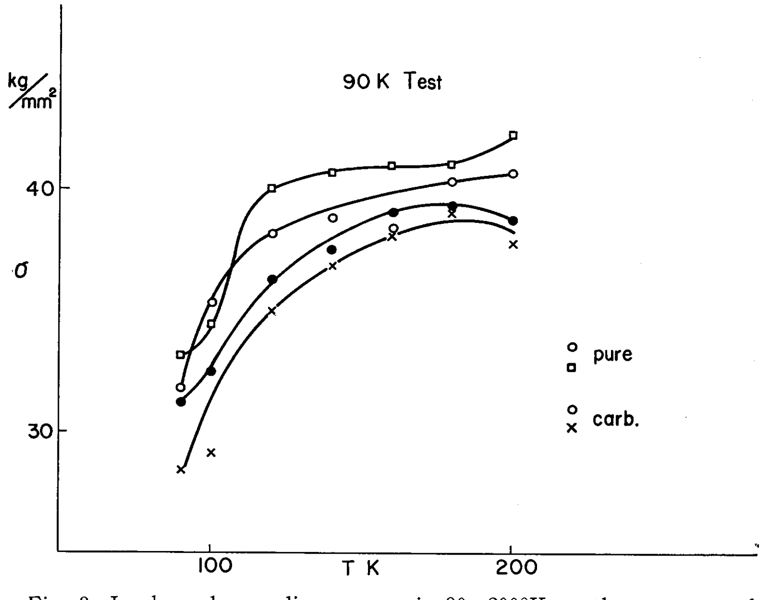


Fig. 3 Isochronal annealing curves in 90~200°K on the recovery of 90°K yield stress of pure iron and iron containing 10 ppm C single crystals with same orientation. 20K/30min.

annealing curves near 370 °K are shown in Figs. 8 and 9 for pure iron and iron containing large amount of carbon respectively. Fig. 10 summarizes the recovery characteristics of single crystals of pure iron and iron containing large amount of carbon over whole range of temperatures, where the chain lines show the results obtained for pure iron and iron containing carbon irradiated at ambient temperature.

Fig. 11 shows the temperature dependence of flow stress in single crystals of pure iron as irradiated at low temperature, and after annealed at 200 °K comparing with those in unirradiated pure iron, and pure iron and iron containing large amount of carbon (100 ppm) irradiated at ambient temperature.

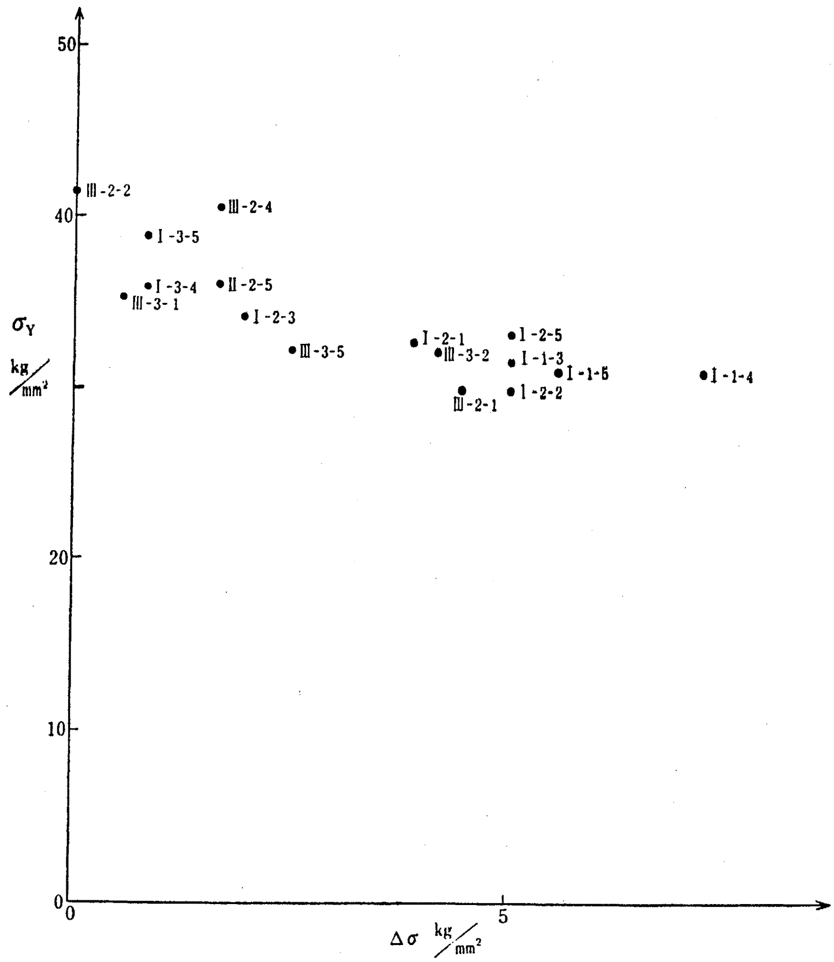


Fig. 4 Increments of 90°K yield stresses by annealings up to 200°K $\triangle\sigma$ vs. 90°K yield stresses of pure iron and 50 ppm C-iron single crystals, I-3-4, 5, III-2-1, 2, 4 are 50 ppm C-iron.

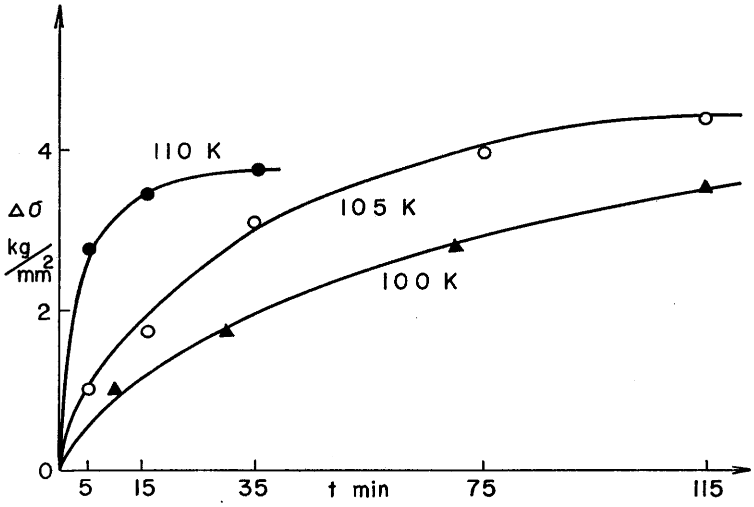


Fig. 5 Isothermal annealing curves at 100, 105 and 110°K on the recovery of 90°K yield stress of pure iron single crystals with same orientation.

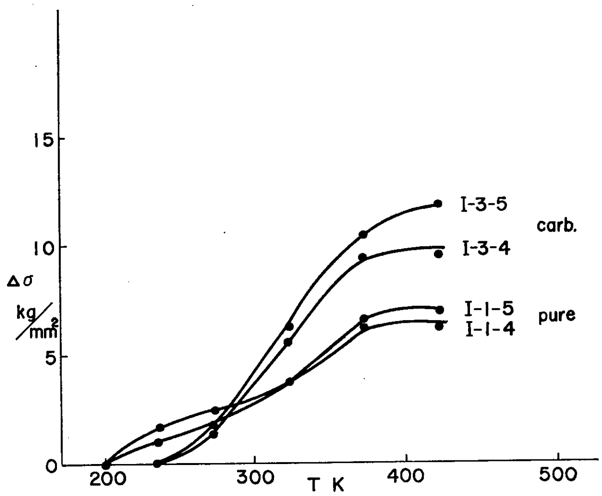


Fig. 6 Isochronal annealing curve in 200~420°K on the recovery of 200°K yield stress of pure iron and 50 ppm C-iron single crystals with same orientation.

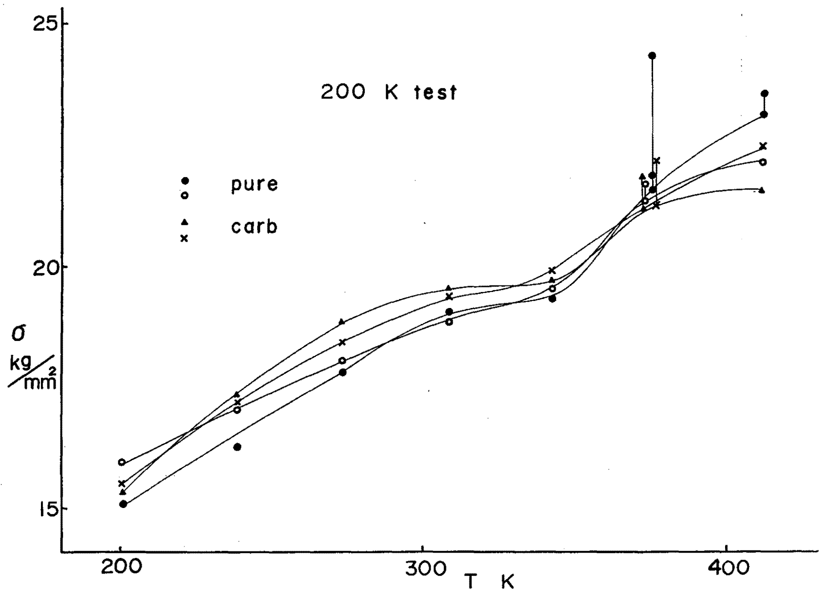


Fig. 7 Isochronal annealing curves in 200∼420°K on the recovery of 200°K yield stress of pure iron and 10 ppm C-iron single crystals with same orientation.

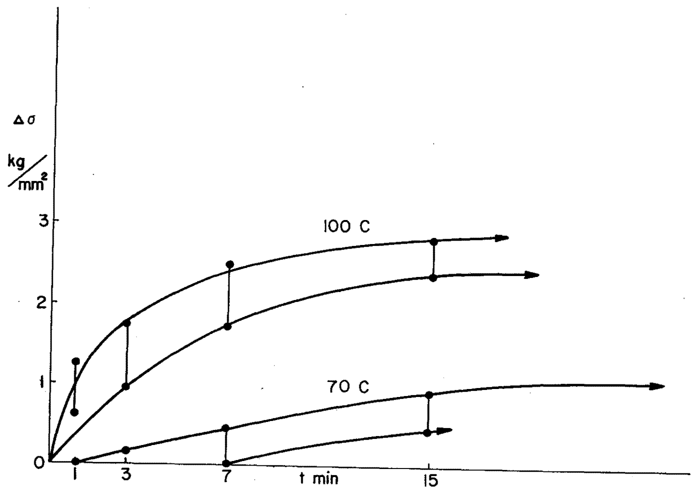


Fig. 8 Isothermal annealing curves at 343°K and 373°K on the recovery of pure iron single crystals with same orientation.

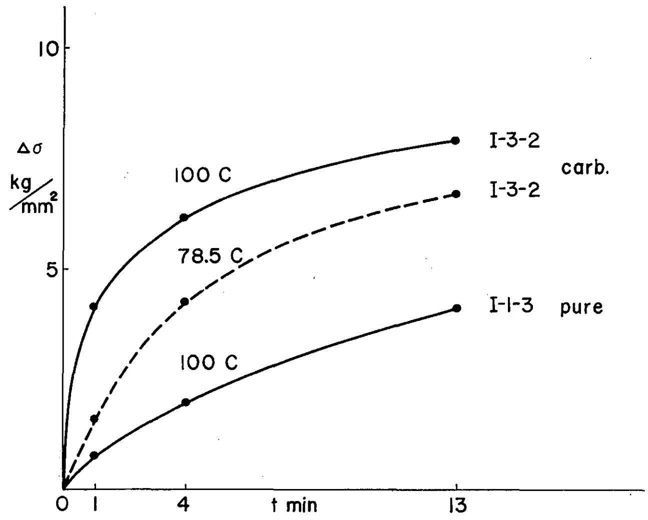


Fig. 9 Isothermal annealing curves at 351°K and 373°K on the recovery of 10 ppm C-iron single crystals with same orientation.

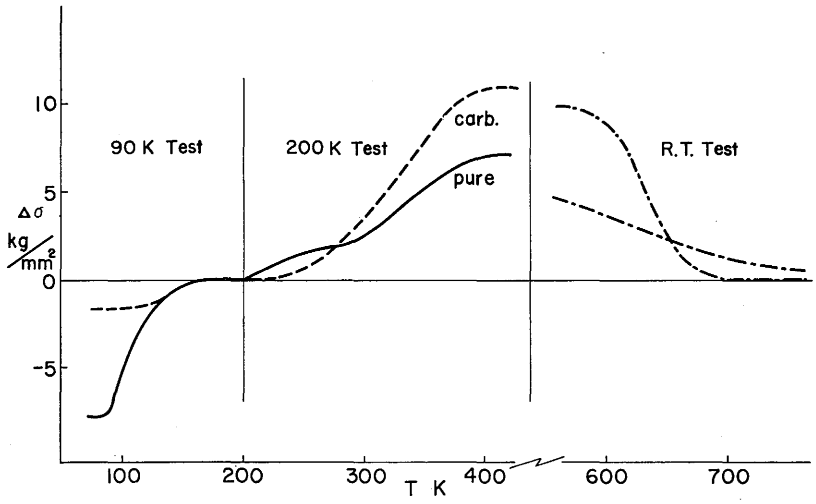


Fig. 10 General characteristics of isochronal annealing curves of pure iron and 50 ppm C-iron single crystals irradiated at low temperature. 90°K test for 90～200°K, 200°K test for 200～420°K. 300°K test for above 420°K on ambient temperature irradiated specimen.

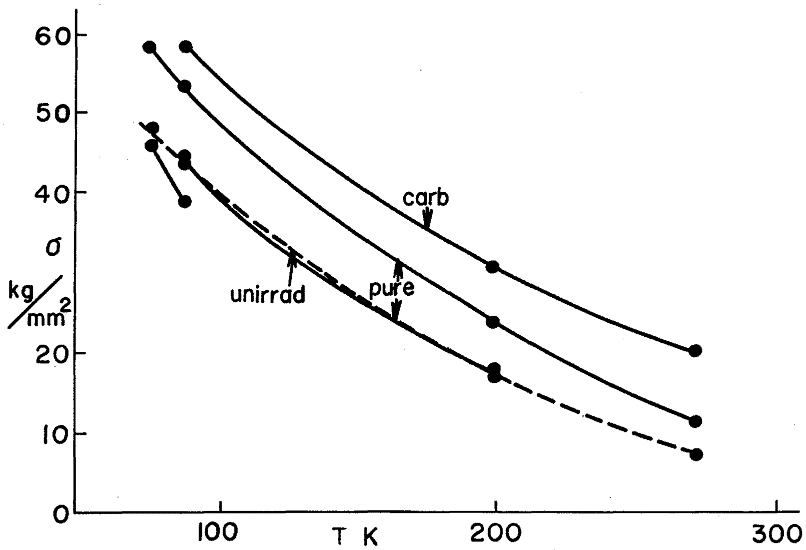


Fig. 11 General characteristics of temperature dependence of yield stresses of irradiated iron single crystals. The low temperature irradiated pure iron, the one annealed up to 200°K, the ambient temperature irradiated pure iron, and the iron containing 100 ppm C for full lines upward from lower most to upper most curves. respectively Dotted curve is unirradiated specimen.

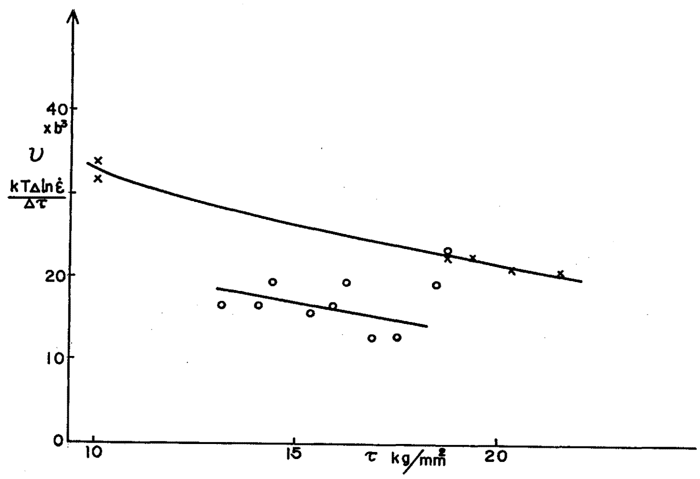


Fig. 12 The stress dependence of activation volume of low temperature irradiated pure iron single crystal, open circle, and the one annealed at 200°K, cross mark.

We may notice that the softening at 77 °K is smaller than that of 90 °K. This tendency was confirmed on many other specimens. Temperature dependent hardening and temperature independent hardening in single crystals of pure iron and iron containing large amount of carbon were reported in the previous paper [12]. Fig. 12 shows the comparison of activation volumes in irradiation softened single crystals of pure iron and the same crystals annealed up to 200°K. The activation volume is smaller in softened crystals, while that in recovered crystals is nearly equal to that of unirradiated ones.

## 4. Discussion

Softening observed at the temperatures $<90°K$ in irradiated iron may correspond to those in electron irradiated specimens reported by Meshii [2)4)5)] (E=2 MeV, $10^{16} \sim 5\times10^{18}$ e/cm$^2$, $<60°K$) and Groh [6)] (E=3 MeV, $10^{17} \sim 5\times 10^{18}$ e/cm$^2$, 20°K) and in neutron irradiated specimens reported by Groh [6)] (E>1 MeV, $5\times10^{15} \sim 5\times10^{18}$ n/cm$^2$). Comparisons of the amount of softening among these data, however, are difficult because test temperatures, specimens used and types of irradiation are diverse. Specimens of Groh are polycrystals of 0.2 mm thickness plate with grain size of 0.01 mm, while those of Meshii are single crystals of $0.3\times1.2\times3$ mm and $0.35\times1.2\times7$ mm. Following to Groh [6)] we shall assume the production of Frenkel pair is $10^{-16}$ per e/cm$^2$ for electron irradiation, while $5\times10^{-16}$ per n/cm$^2$ for neutron irradiation, then our $3\times10^{16}$ n/cm$^2$ corresponds to $1.5\times10^{17}$ e/cm$^2$. Meshii's data show $\Delta\sigma=2$ kg/mm$^2$ for tests at 60°K by $2\times10^{17}$ e/cm$^2$. This is rather small compared with $\Delta\sigma=5$ kg/mm$^2$ for test at 90°K. This may partly due to the difference of test temperatures. Groh however showed $\Delta\sigma=35$ kg/mm$^2$ for $3\times10^{18}$ e/cm$^2$ and $\Delta\sigma=20$ kg/mm$^2$ for $7\times10^{17}$ n/cm$^2$, and stated that homogeneously distributed defects obtained by electron irradiation produce larger softening then inhomogeneous ones by neutron. Furthermore the temperature dependence of yield stress in the softening is different, i.e. Meshii's data show softening is maximum about$\sim$60°K and decreases as the temperature increases, in contrast to our case where softening increases up to 90°K, Fig. 11. On the other hand Groh's data show softening is minus for $<6\times10^{16}$ n/cm$^2$, while ours $\Delta\sigma=2$ kg/mm$^2$ for$\sim1\times10^{16}$ n/cm$^2$, and softening decreases from 77°K to 100°K contrary to ours. These discrepancies among data must be re-examined by further works.

Meshii [3)] proposed a theory for softening phenomena in BCC metals containing dispersed lattice defects. He assumed the linear elastic interaction between defects and dislocations and showed that when Peierls stress of screw dislocation is large, the part of dislocation near the defects is bowed by the stress field of the defects, thus the formation of kink pair is assisted. His theory may be reasonable when the mean distance is larger than the size of kink pair, but it may be difficult to explain similar softening in the case mean distance is smaller. Precise nature of neutron irradiated defects is yet unknown at the present time but available data show that the size of depleted zone is about 100 Å and about 100 interstitials associate to one depleted zone. Then the mean distance of interstitials may be about 5 atomic spacings. On the other hand the size of kink pair is about 20 atomic spacings according to Fig. 12.

Another explanation may be to take into account the interaction between dislocation core and defect. According to Suzuki's theory[14], the large Peierls stress of screw dislocation in BCC metal is attributed to the arrangement of atoms in the core of screw dislocation peculiar to BCC lattice, *i. e.* equally spaced screw arrangement of atoms in the three neighbouring atomic rows along the axis $\langle 111 \rangle$ in perfect lattice without dislocation changes to similar screw arrangement inside the core of screw dislocation where core potential is low, but changes to enforced symmetrical arrangement when the screw dislocation moves one atomic distance, where the core potential is high. Such cyclic change of potential of the core of screw dislocation in the movement is the main cause of large Peierls potential. Then any cause which disturbs the regularity of the lattice may lower the Peierls potential locally, and formation of kink pair may be assisted. In this view both interstitial and vacancy are able to cause softening. A more accurate description, of course, must wait atomistic calculations. The decrease of activation volume by softening as shown in Fig. 12 is not inconsistent with the present explanation.

Then the recovery of softening near 100°K in Fig. 2 may correspond to those of electric resistivity and internal friction in I stage[1]. From Fig. 3 activation energy is calculated as 0.26~0.27 eV which coincides with the value $0.26 \pm 0.03$ eV reported by Bilger[1] for $I_E$ stage recovery. We may notice here the recovery is rather sharp compared with Groh's one[6].

From Fig. 5. we can see the two recovery stages in 160~320°K and 320~470°K. The latter may correspond to the hardenings seen in specimens irradiated at ambient temperature. The defects responsible for the hardening may be small cluster of interstitials formed by movement of some defects in these stages. But we can not identify them at present.

According to observations[13] by electron microscope the defects produced by ambient temperature irradiation up to $10^{19}$ nvt (>1 MeV) do not produce any image contrast. So we may surmise that the size of defects may be as small as $\lesssim$10 Å.

The iron containing large amount of carbon produces larger hardening than the pure iron as shown in Fig. 6. And the activation energy of the process of hardening is about 0.8 eV from Fig. 9. which is nearly equal to that of diffusion of carbon atoms. From these evidences we may deduce that the hardening is caused by the complex defects composed of interstitial clusters combined with carbon atoms. The rapid recovery of the hardening in the stage near 300°C in Fig. 8 may be due to the dissociation of carbon from the complex clusters[12].

The interpretation for the temperature dependence of yield stresses of irradiated single crystals shown in Fig. 11 is as follows; low temperature irradiated specimen shows softening of yield stress caused by interaction of dispersed interstitials with screw dislocation, the softening is recovered in the specimen annealed up to 200°K, where interstitials were disappeared by meeting to vacancies or forming small cluster of interstitials. The clusters, then, grow to larger one by migration of clusters at 100°C and cause hardening, which is weak and temperature dependent since the cluster is rather small size and not stable in the interaction with dislocation. However the clusters may be grown to larger one and stabilized combining with carbon atoms near 100°C, and form firm obstacles against the movement of screw dislcations which result in the tempereture independent type hardening as shown in iron containing large amount of carbon.

**Acknowlegements**

The authors would like to present their cordial thanks to Messrs. T. Fujiwara and Y. Miyamoto for their assistances in experimental works, to members of Committee of Common Use of Low Temperature Irradiation Loop of KUR in Kyoto University for their co-operations in lavour of irradiation and valuable discussions. This work is founded on the Inter-Uinversities Comon Use Program of Facilities in Research Reactor Institute of Kyoto Uuiversity.

**Reference**

1) Bilger, H., Hivert, H., Verdone, J., Leveque, J.L. and Soulie, J.C., Point Defectsi n Iron, *Proc. Intern. Conf. Vacancies and Interstitials in Metals. II,* Kernforschungsanlage, Jülich, 1968, p. 751.
2) Sato, A., Mifune, T., Meshii, M., Effect of Electron Irradiation on the Strength of Iron Single Crystals, Proc. *2nd Int. Conf. Strength of Metals and Alloys. II,* Am. Soc. Metals, 1970, p. 747.
3) Sato, A., Meshii, M., Solid Solution Softening and Solution Hardening, Acta Met., Vol. 21, 1973, p. 753.
4) Sato, A., Mifune, T., Meshii, M., Irradiation Softening in Pure Iron Single Crystals, Phys. Stat. Sol. (a), Vol. 18. 1973, p. 699.
5) Sato, A., Meshii, M., Asymmetry in Yield Stress and Irradiation Softening of High Purity Iron Single Crystals, Scripta Met., Vol. 8, 1974, p. 851.
6) Groh, G., Vanoni, F., Moser, P., Low Temperature Effects on Deformation Characteristics in Iron, *Point Defects and their Clusters in BCC Metals and Alloys,* Ed. Arsenault, Nucl. Metull, Vol. 18, 1973, p. 19.
7) Kitajima, K., Radiation Effects in Iron Single Crystals, *U.S.-Japan Seminar on Radiation Effects in Metals and Structural Materials,* Kyoto, 1971.
8) Kitajima, K., Abe, H., Low Temperature Neutron Irradiation Effects on Iron Single Crystals, Bulletin Res. Inst. Appl. Mech., Kyushu Univ., No. 40, 1973. (in Japanese)
9) Chow. J.G.Y., McRickard, S.B. and Gurinsky, D.H., Mechanical Properties of Irradiated Iron and Iron Alloys, *Radiation Effects on Metals and Neutron Dosimetry,* ASTM STP 341, Am. Soc. Testing Mats., 1962, p. 46., Chow, J.G.Y. and McRickard, S.B., Low Temperature Embrittlement of Iron, Iron Alloys and Steels by Neutron Irradiations, *Flow and Fracture of Metals and Alloys in Nuclear Environments,* ASTM STP 380, Am. Soc. Testing Mats., 1964. p. 120.

10) McRickard, S. B. and Chow, J. G. Y., The Effect of Interstitial Impurities on Radiation Hardening and Embrittlement in Iron, Acta Met., Vol. 14, 1966, p. 1195.
11) Ohr, S. M., Hinkle, L. E., Wechsler, M. S. and Chen, C. W., Radiation Hardening and Radiation-Anneal Hardening in Body-Centered Cubic Metals, *Proc. 2nd Intern. Conf. Strength of Metals and Alloys*, Am. Soc. Metals, 1971, p. 742.
12) Kita jima, K., Neutron Irradiation Effects on the Plastic Properties of Pure Iron, *Proc. Intern. Conf. Strength of Metals and Alloys*, Supplement Trans. Japan Inst. Metals, Vol. 9, 1968, p. 182.
13) Bryner, J. S., A Study of Neutron Irradiation Damage in Iron by Electron-Transmission Microscopy, Acta Met., Vol. 14, 1966, p. 323., Bryner J. S. and Chow, J. G. Y., Fast-Neutron Irradiation Effects on Dislocation Structures in Iron Strained at Ambient Temperature, Acta Met., Vol. 18., 1970, p. 191.
14) H. Suzuki, Motion of Dislocation in Body-Centered Cubic crystals, *Dislocation Dynamics*, (McGraw-Hill, 1968, New York), p. 679.

(Received December 26, 1974)