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NEUTRON IRRADIATION EFFECTS ON THE PLASTIC PROPERTIES OF PURE IRON

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Stress-strain characteristics together with internal frictions were measured on pure iron single crystals containing < 0.1, 1 and 20 ppm carbon and irradiated to doses of $0, 1.8 \times 10^{17}$, 1.3×10^{18} and 1×10^{19} nvt (>1 MeV) at temperature $\sim 70°C$. The increases of yield stress at small strain and subsequent flow stress at larger strain were interpreted as caused mainly by barriers of irradiation defects against the movements of edge and screw dislocations respectively based on the standpoint that the latter has much larger Peierls stress than the former. Appreciable decrease of activation volume associated with the flow stress observed in irradiated specimens was attributed to the process of screw dislocation overcoming Peierls hill near the top of irradiation defects of sufficiently small size, presumably such as triplet of interstitials or small dislocation loop invisible by electron microscope. The increase of the flow stress estimated from the present view are shown to coincide with the observed value.

The contents of carbon were found not to have large effects on hardening in as irradiated condition within the dose of $< 1.3 \times 10^{18}$ nvt, though they gave large effects in the course of recovery of yield stress and internal friction after annealings. The explanations are presented for these evidences based on the mechanisms of trapping and detrapping of carbon atoms to vacancies and other defects during annealing.

1. Introduction.

Compared with those in FCC metals the studies for hardening by neutron irradiation in iron have met with difficulties on the two sides, the one concerns with the fundamental mechanisms of plastic deformation in unirradiated pure iron, and the other the nature of latice defects produced by irradiation.

Recent experimental studies [1)~6)] on the former problem, clarified the main features of plastic deformation in BCC metals, though there remain some ambiguities yet in the interpretations of mechanims. On the other hand, there are reported many observations by electron microscope on the nature of defects in irradiated BCC metals [7)~9)] according to which defect clusters of invisible size are considered to be mainly responsible for harden-

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ing. Important rôle of interstitial impurities on the structure of defects were pointed out by many authors [7)~12)]. The rôle of these defects and impurities in the mechanism of hardening, however, are not yet fully separated from the characteristics in unirradiated pure iron, because the measurements made so far are rather confined to poly-crystals [13)].

In this paper to solve the above problems the author provides some data from the measurements of stress-strain curves and internal frictions of single crystals of very pure iron and iron containing small amount of carbon, including the effects of annealing.

II. Specimens and Procedure of Experiments.

Starting materials were carbonyl iron GA & F J which were spectroscopically pure in metallic impurities except O, C and N. Pure iron was produced from the material by zone-refining with 6 passes in pure hydrogen and one pass in vacuum. Single crystals were made by strain anneal method after nitrizing them by 0.005 % N, which prevent easy recrystalization. Then they were denitrized by wet hydrogen treatment at 720°C for 50hr [5)]. Interstitial impurities remained, mainly carbon, were about $0.5 \sim 1$ ppm. some of the speciments were then purified further by annealing them for 14 days at 880°C in enclosed ZrH_2 purifying circuit [3)]. By this treatment carbon contents were estimated to have been reduced to < 0.1 ppm. And some of the samples were re-carburized so as to give them 20 ppm carbon content and quenched after homogenizing.

Irradiations were performed in the reactor KUR of Kyoto University to the doses of 1.8×10^{17} and 1.3×10^{18} nvt (> 1 MeV) at the irradiation tempetature 60°C $\sim$ 90°C. Specimens of ~ 1 ppm were also irradiated in the reactor BR-2 of EURATOM-CEN Belgium to dose of 1×10^{19} nvt (> 1 MeV) at the temperature controlled to 75 ± 10°C.

Tensile tests were performed on specimens of $1 \times 5 \times 50$ mm at temperatures of 295° $\sim$ 77° K and strain rate dependency of flow stress was measured by instantaneous change of cross head speed. Isochronals for room temperature yield stress were measured over the range of 100°C to 700°C. On the other hand, internal frictions were measured on specimens of $1 \times 5 \times 90$ mm in the magnetic field of 200 Oe by the method of lateral vibration, in which effects of cold work and isochronal annealings were also included.

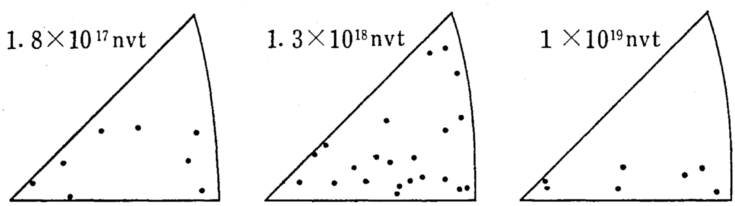


Fig. 1. Orientations of single crystals used in this experiments.

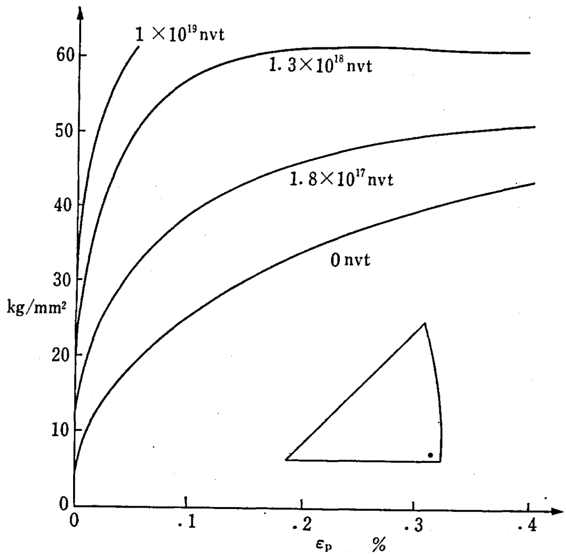


Fig. 2. Dose dependence of stress-strain curves at 77°K in pre-strained single crystals with same orientation.

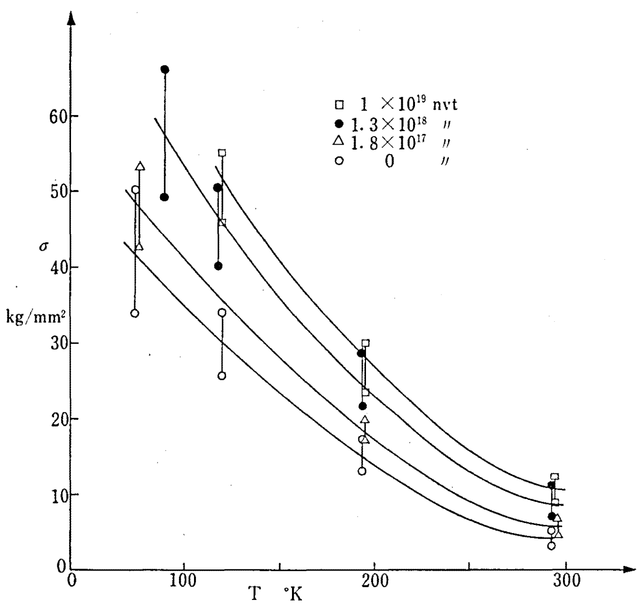


Fig. 3. Dose and temperature dependence of saturated flow stresses of single crystals. Range of experimental Points and curves for representative values are shown.

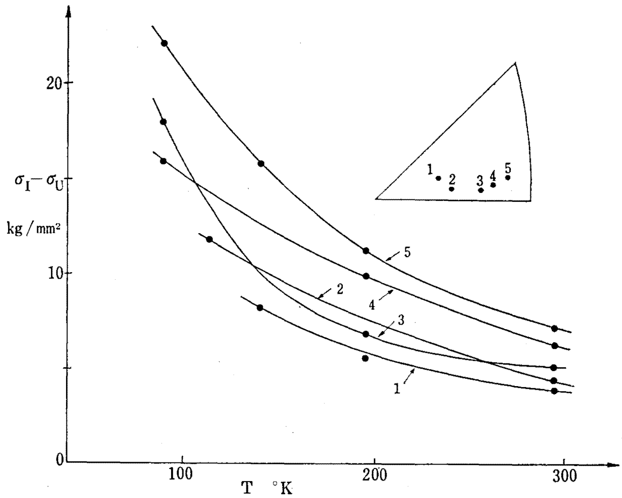


Fig. 4. Differences between flow stresses of irradiated and unirradiated single crystals of same orientations, No. 5; 1×10^{19} nvt, others; 1.3×10^{18} nvt.

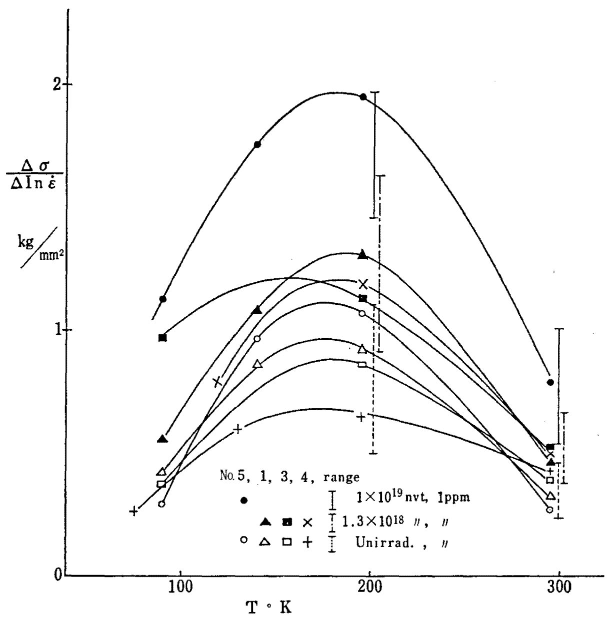


Fig. 5. Comparison of strain rate sensitivity. Number of specimens are same with Fig. 4.

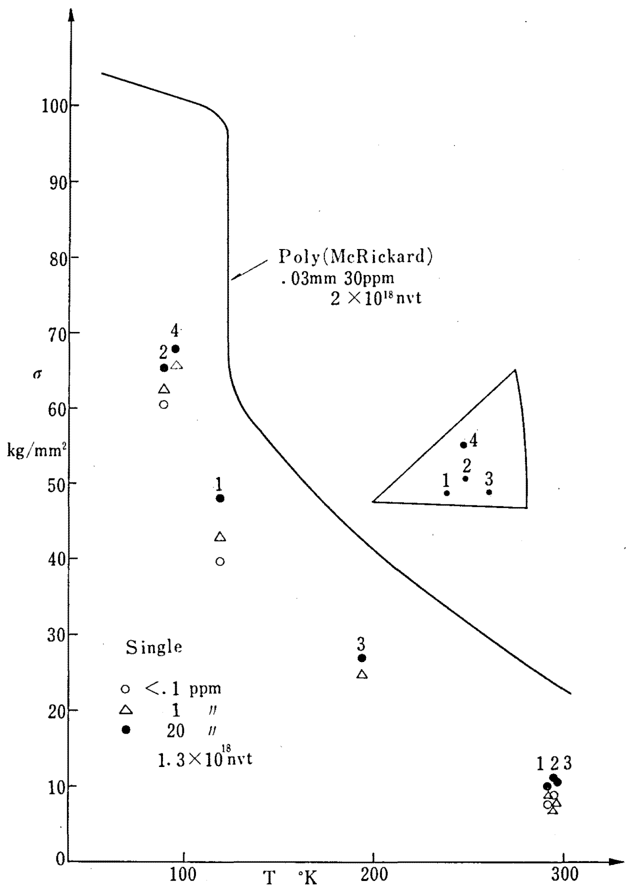


Fig. 6. Comparisons of flow stresses in single crystals which have various carbon contents.

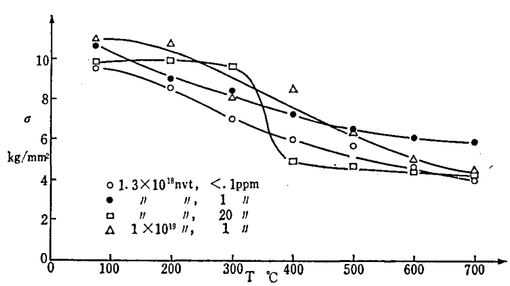


Fig. 7. Isochronals (30min/100°C) of room temperature yield stress in irradiated single crystals.

III. Experimental Results

1. Tensile tests

Orientations of the single crystals used in the experiment are shown in Fig. 1. Specimens were pre-strained first about 0.3 % at room temperature, then stress-strain curves were measured at lower temperatures [4)~6)]. Fig. 2 shows an example tested at 77° K on the single crystals with the same orientation. The yield stress together with the rate of work-hardening increases with the increase of dose. The flow stresses are then saturated to nearly constant values in the range of larger strains, whose temperature and dose dependencies are shown in Fig. 3 on specimens of various orientations.

On the other hand, Fig. 4 shows differences of the flow stress between irradiated and unirradiated specimen which have same orientations. Strain rate dependency of the flow stresses on these specimens are also shown in Fig. 5. We can see that the difference of flow stress depends upon temperature, and the strain rate sensitivity is appreciably larger in irradiated specimens. In general twining is suppressed, not observed in $\geq 1.3 \times 10^{18}$ nvt and $\lesssim$ 1 ppm specimens, and transision temperature is increased by irradiations.

The flow stresses increased as carbon content increased from < 0.1 ppm to 1 ppm and 20 ppm on 1.3×10^{18} nvt irradiated single crystals of same orientation, but the differences between them were not so large as seen in Fig. 6. However, remarkable effects of carbon content were observed on the recovery of room temperature yield stress by isochronal annealing (30 min/100°C), Fig. 7, in which the specimens of < 0.1 and 1 ppm C show similar slow recovery in contrast to the sharp recovery near 350°C in specimen of 20 ppm C.

2. Internal friction

Internal friction of the specimens did not show so much change by irradiation except those containing 20 ppm carbon, in which Snoek peak disappeared after irradiation and reappeared after annealing at the temperatures higher than 250°C [11)]. On the other hand, specimens cold worked by 3～4 % at room temperature after irradiation showed characteristic variations as compared with the unirradiated ones. In the irradiated specimens, amplitude dependency of internal fiction is much smaller in as cold worked state, and rather stable peaks appeared more frequently after appropriate annealings, Figs. 8, 9, 10. However, characteristics of the peaks are rather compricated in common with the case of unirradiated specimens [14)], so that details will be reported in other works. Fig. 11 summarizes the effects of isochronal annealing (15 min/20°C) on the room temperature internal friction in the as cold worked specimens of various carbon contents. Recovery temperatures of 20, 1 and < 0.1 ppm specimens are shifted by irradiation to higher temperatures of

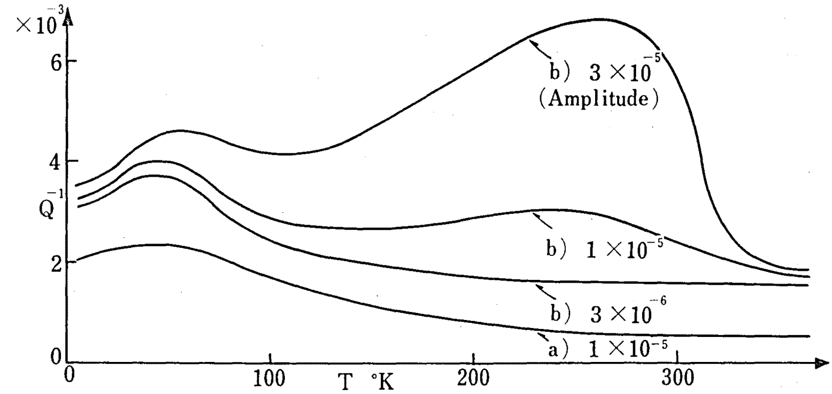


Fig. 8. Internal friction of unirradiated lppm specimen.
a) As annealed, b) 3% cold worked.

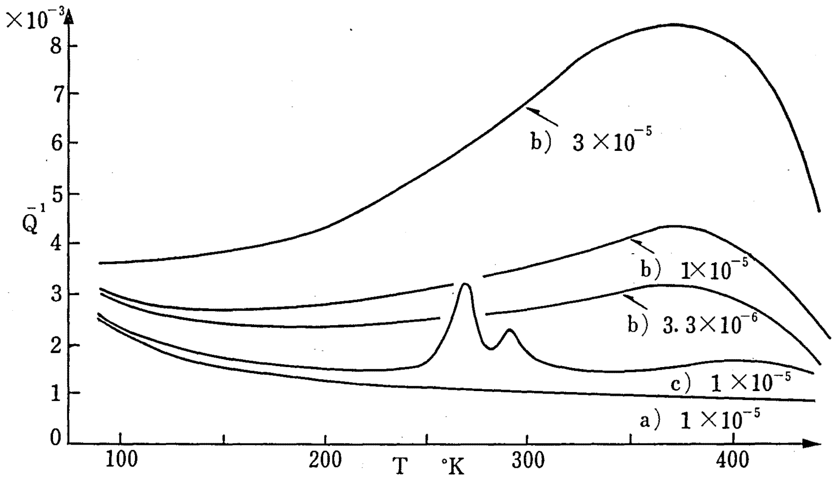
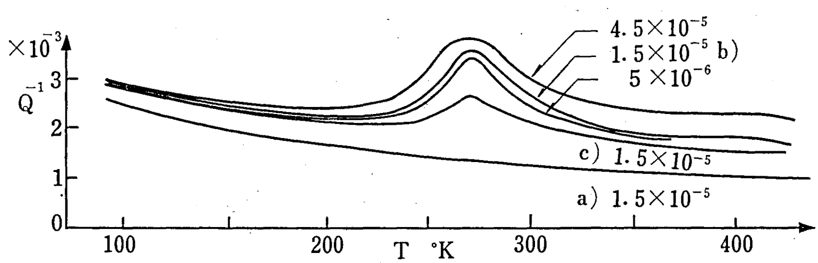


Fig. 9. Internal friction of 0.1ppm, unirradiated specimen,
a) As annealed, b) 3% cold worked, c) Annealed
30min at 300°C after cold work.



Fg. 10. Internal friction of lppm, 1.3×10^{18} nvt specimen,
a) As irradiated, b) 4% cold worked, c) Annealed
30min at 250°C after cold work.

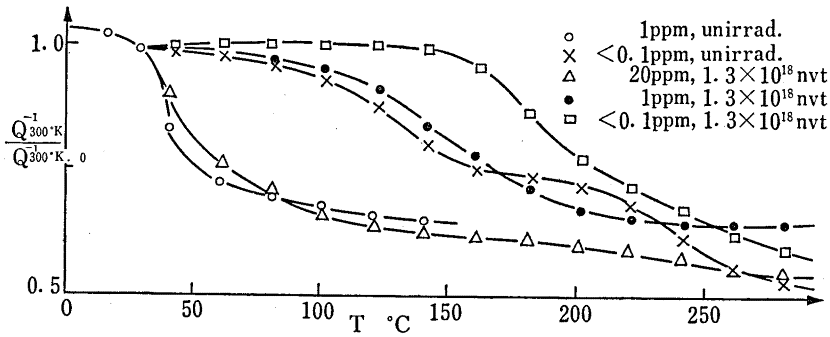


Fig. 11. Effects of isochronal annealing (15min/20°C) on room temperature internal friction of 3% cold worked specimens.

about 70°, 150° and 250°C respectively.

IV. Discussion

Difficulties in reasonable explanations for irradiation hardening in iron are partly related to some ambiguities remained in the fundamental mechanisms in the plastic deformation in BCC metals. The theories presented by Hirsch [15] and Suzuki [16], however, may have clarified some confusions in the opinions [1)~6)], the details of which will be discussed in a later work. According to these theories, screw component of dislocation in BCC metals is predicted to have much larger Peierls stress than that of edge component as an intrinsic charactor related to the crystal structure. From this standpoint increments of yield stress by irradiation in Fig. 2 can be interpreted as the stress required for mobil edge dislocations overcoming the barrier of defects introduced by irradiation. Movable edge dislocations may be formed after pre-straining at room temperature, because the dislocations in BCC metals are considered to contain many jogs [17] in annealed state consequently movable segments are rare even if contents of carbon is very small [3]. These movable edge dislocations may be exhausted after forming single elongated dislocation loops sided by screw dislocations by straining at lower temperatures where screw dislocations are immobile. This exhaustion mechanism may explain the observed rapid hardening in Fig. 2.

On the other hand, the flow sstressess saturated to nearly constant values at larger strains shown in Figs. 3 and 4 may be attributed to further movements of screw dislocations thus formed. Figures 3~5 also show that the process of sorew dislocations overcoming the barrier of irradiation defects is a thermal process instead of the athermal one as suggested by the results in polycrystals [13]. The increments of flow stress at 0°K, $(\sigma_I - \sigma_U)_0$, can be estimated by using the formula of Fleisher [18],

$$(\sigma_I - \sigma_U)_0 = \frac{2}{3.3} G (bc/d)^{\frac{1}{2}} \quad ,$$

where G is shear modulus, b the Burgers vector, c the atomic concentration of defect forming dislocation loops d the diameter of the loop. If we assme $b/d = 1/2 \sim 1/4$ for the size of clusters by the reasons will be stated later, and $c = 1/2 \times 10^{-3}$ by the reasoning that the cocentration of primary Frenkel pair introduced by irradiation is estimated to be $1/2.5 \times 10^{21}/\mathrm{cm}^3$ for dose of 1×10^{19} nvt following to Eyre [7], and further one tenth of which survive as clusters at the present irradiation temperature, then we obtain $(\sigma_I - \sigma_U) = 75 \sim 53\ \mathrm{kg/mm^2}$. This is right order of magnitude compared with the experimental value of $(\sigma_I - \sigma_U)_o = 30\ \mathrm{kg/mm^2}$, which is obtained exterpolating the curve 5 in Fig. 4 by means of the formula of Fleischer [18],

$$(\sigma_I - \sigma_U)_T/(\sigma_I - \sigma_U)_o = \left\{1 - \left(\frac{T}{T_o}\right)^{\frac{1}{2}}\right\}^2 \quad .$$

However, the activation process is more complicated compared with those in FCC metals [19], because it is considered to be a multiple activation energy process in the meaning that the large Peierls hill is superposed on the stress field around the defects in the case of BBC metals. Activaticn volume defined by $2kT \triangle ln \dot{\varepsilon} / \triangle\sigma$ is appreciably smaller in the case of irradiated specimens than those of unirradiated specimens as shown in Fig. 5. Such a marked decrease of activation volume may be attributed to sufficiently small size of barrier introduced by irradiation, since rather larger activation volume was observed in precipitation hardened iron [2] and similar value of activation volume was observed in latent hardening caused dy multiple slip [20]. In fact, at the cusp of bowed dislocations near the top of barrier the activation volume associated with the process overcoming Peierls hill may be reduced appreciably if the size of barrier is so small as comparable with the breadth of thermal kink pair. On the other hand, rather large values of activation energy and activation volume similar to the cases of FCC metals [19] might be expected on the process overcoming the stress field around the barrier, but the effects of them is considered to be masked in the observed strain rate sensitivity by the variation of activation volume in the former process, *i. e.* that of overcoming Peierls hill. The explanations seem to coincide with the observations by electron microscope, that is, the size of defects responsible for most of hardening is invisible one and presumably smaller than 10Å in our radiation condition [7][8]. Entities of such defects may be dispersed clusters of frozen vacancies, as suggested in Seeger's depleted zone [21], and stable clusters of interstitials, say triplet [9] or small dislocation loop, formed by recombination of interstitials during irrdiation [7].

Fig. 6 shows that small quantity of carbon does not affect appreciably the properties of defects responsible for hardening at least at low temperatures, though it may have important rôle in other properties of the defects through trapping to interstitials and vacancies [10][11]. McRickard and Chow [12] reported that the 30 ppm carbon introduced abrupt chages in hardening and fracture characteristies of pure iron poly-crystals at $< 120°$K by room temperature irradiation of 2×10^{18} nvt (> 1.45 MeV), Fig. 6. The discrepancy might be attributed to characteristics of poly-crystals or still lower dose in our experiments.

However, the effects of carbon is appreciable in the process of recovery of room temperature yield stress by annealings, Fig. 7. Since all carbon atoms are considered to be trapped to single vacancies[10] and other defects in our irradiation condition, the sharp recovery of yield stress near 300°C in 20 ppm specimen, as shown in Fig. 7, may be attributed to detrapping of carbon atoms from vacancies near 250°C[10] and subsequent migration of vacancies near 300°C[10]. In contrast to it, slower recovery in ~ 1 ppm and < 0.1 ppm specimens may be due to insuffcient numbers of carbon atoms to trap all vacancies, consequently to annihilation of neighboring interstitial clusters and vacancies at lower temperatures. The reason why recovery is incomplete at 300°C compared with 20 ppm specimen is not clear at present stage.

Internal friction of cold worked unirradiated specimen of ~ 1 ppm C shows a typical broad peak near room temperature, as shown in Fig, 8. From the fact that the peak tempedature shifts to higher side in < 0.1 ppm specimen as seen in Fig. 9, and from the recovery kinetics of the room temperature internal friction as shown in Fig. 11, we can interpret that the decrease of internal friction by annealings is attributed to the pinning of dislocations by carbon atoms. Then the delay of recovery in irradiated specimens as shown in Fig. 11 can be reasonably interpreted by the trapping of carbon atoms to vacancies and other defects introduced by irradiation and the detrapping of them by annealings at higher temperatures. However, in contrast to the recovery of yield stress, Fig. 11 shows that detrapping temperature varies among 20, 1 and < 0.1 ppm specimens. This may be due to the varieties of trapped state of carbons to various defects, whose effects are exaggerated in the case of internal friction since very small amount of untrapped carbon atoms is considered to be sufficient to pin the dislocations.

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