

DISCUSSION

*F. A. Smidt, Jr.*¹—The authors have added some valuable knowledge to our understanding of the effects of alloying elements on irradiation hardening of ferrous materials containing nitrogen in solution during 60 C (140 F) irradiations. However, their identification of the hardening mechanism as “nitrogen bound by irradiation-induced point defects” or “complex defects” is contrary to the conclusion of numerous other workers in the field [8,9,10]^{2,3} that irradiation hardening in this temperature range is produced by the precipitation of the interstitial solutes on dislocation loops formed during the irradiation. This process converts these loops from relatively soft barriers to dislocation motion, approximately one half the strength of a precipitate, to hard precipitatelike barriers. Among the works contributing to this understanding were the demonstration by Bullough, Stanley, and Williams² that trapping of oxygen in columbium by a defect produced during the irradiation was consistent with trapping at dislocation loops. Wuttig, Stanley, and Birnbaum³ investigated the analogous problem for carbon and nitrogen in iron, using magnetic disaccommodation to study the trapping mechanism in irradiated and quenched materials, and concluded that the trapping sites during low-temperature irradiations were the dislocation loops produced during irradiation. Ohr et al [8] summarize a number of observations of radiation hardening in iron, columbium, and vanadium that are consistent with precipitation of interstitial impurities on dislocation loops. Smidt [9] has shown that anneal hardening in vanadium results from a similar mechanism. Stanley et al [10] have also observed the phenomenon in vanadium, and Little and Harries⁴ note the likelihood of its occurrence for nitrogen in mild steel. Since there appears to be no contrary evidence in the current investigation, trapping of nitrogen at dislocation loops would appear to be the operative hardening mechanism in this case too. The

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² Bullough, R., Stanley, J. T., and Williams, J. M., *Metal Science Journal*, Vol. 2, 1968, p. 93.

³ Wuttig, M., Stanley, J. T., and Birnbaum, H. K., *Physica Status Solidi*, Vol. 27, 1968, p. 701.

⁴ Little, E. A. and Harries, D. R., “Effects of Interstitial Elements on Radiation Hardening in Mild Steels,” *Irradiation Effects in Structural Alloys for Thermal and Fast Reactors*, ASTM STP 457, American Society for Testing and Materials, 1969, p. 215.

annealing of irradiation hardening and the reappearance of precipitates during postirradiation annealing are also consistent with the release of nitrogen from small dislocation loops observed by Wuttig et al.³ to take place at 200 C (392 F).

Specific exceptions are taken to the interpretation of embrittlement in copper-bearing alloys as due to the aforementioned mechanism. First, the authors should note that the presence of copper produces enhanced irradiation hardening in a pure zone-refined, hydrogen-purified material free of nitrogen.⁵ Also, the temperature dependence of their proposed mechanism is wrong since copper does not increase embrittlement at irradiation temperatures of 60 C (140 F) but does for irradiation at 290 C (554 F). It should be noted that the trapping of nitrogen at loops that produces hardening in the lower temperature irradiations would not occur in the high temperature (290 C) (554 F) irradiations since the temperature is above that at which nitrogen is released from the traps (200 C) (392 F) in Wuttig's experiment.³ Finally, observations of the true defect microstructure by TEM require magnifications considerably higher than those employed in this investigation.⁵

N. Igata (authors' closure)—The identification of the essential defects for postirradiation anneal hardening [8,9] a little above Stage III, which would correspond to the case of hardening of iron irradiated at 60~70 C (140~158 F) [2], seems not to be settled [9].⁴ The two mechanisms would be those of obstacle hardening due to interstitial impurity atoms trapped by visible interstitial dislocation loops [8], and invisible defects, possibly vacancies or vacancy clusters [2].^{6a, 7} There may be the possibility of the former case in columbium or vanadium [8,9,10],²⁻⁵ but the problems remain for the possibility of the latter case in iron, in which the solubility of interstitial solute atoms, especially oxygen, is much less than in columbium or vanadium; interstitial dislocation loops would also be less because oxygen atoms are essential for forming dislocation loops.^{7(b)} In the case of iron there are data that suggest as follows the mechanism due to "the nitrogen (or carbon) bound (or trapped) by irradiation-induced point defects or defect clusters" or "complex defects":

1. With TEM it is difficult to distinguish between point defects and small dislocation loops.

2. In the case of iron irradiated at 70 ± 10 C (158 ± 18 F), dislocation loops or black dots were not observed corresponding to irradiation hardening below the total flux of 1×10^{19} neutrons (n)/cm².⁸ Other similar results have been reported.^{6(a, b)}

⁵ Smidt, F. A., Jr. and Sprague, J. A., "Property Changes Resulting from Impurity-Defect Interactions in Iron and Pressure Vessel Steel Alloys," *Effects of Radiation on Substructure and Mechanical Properties of Metals and Alloys*, ASTM STP 529, American Society for Testing and Materials, 1973, pp. 78-91.

⁶ Bryner, J. S., (a) *Acta Metallurgica*, Vol. 14, 1966, p. 323; (b) BNL 954 (S-68), Brookhaven National Laboratory Reports, Upton, N. Y., 1965, p. 153.

⁷ (a) McRickard, S. B. and Chow, J. G. Y., *Acta Metallurgica*, Vol. 14, 1966, p. 1195; (b) Chow, J. G. Y. and McRickard, S. B., *Flow and Fracture of Metals and Alloys in Nuclear Environments*, ASTM STP 380, American Society for Testing and Materials, 1965, p. 120.

⁸ Hasiiguti, R. R., Igata, N., Kaneko, H., Kitajima, K., Koda, S., Mima, G., and Yamane, T., to be published.

3. There is the evidence of positron-annihilation experiments, which show that solute carbon (or nitrogen) atoms move to single vacancies or maybe vacancy clusters in electron-irradiated or cold-worked iron.⁹

4. There is the evidence of internal friction, which suggests the relaxation of carbon (or nitrogen) trapped by vacancies in neutron-irradiated [4]¹¹ or cold-worked iron.¹¹ The new peak is different from the interaction peak due to solute carbon (or nitrogen) and dislocations.

5. Depleted zones were observed by field-ion microscopy, and those are estimated to be the origin of the irradiation hardening.¹²

6. McRickard and Chow showed that the defects, which are stable between 203~523 K¹⁸ and possibly vacancy-carbon (or nitrogen) complex are essential in irradiation hardening.^{7(a, b)}

7. In the case of iron-copper when nitrogen is included, the effect of nitrogen would be dominant after irradiation of 3.5×10^{19} n/cm² at 60 C (140 F), and this would be superposed on the interactions of copper and vacancies because if vacancy migration energy is 1.1 eV there would be a possibility of vacancy trap by copper atoms at 60 C (140 F) during several days of irradiation. After postirradiation annealing at 200~300 C (392~572 F) the recovery of nitrogen and copper-nitrogen peak was not observed in internal friction measurements. Accordingly, we suggested that copper atoms also have excess binding energy for solute nitrogen and thus would have a hardening effect.

We would like to continue studies to identify the essential defect more directly.

⁹ Snead, C. L., Jr. and Goland, A. N., *Physical Review*, B, Vol. 3, No. 2, 1971, p. 275.

¹⁰ Wagenblast, H. and Swartz, J. C., *Acta Metallurgica*, Vol. 13, 1965, p. 42.

¹¹ Oren, E. C. and Stephanson, E. T., *Metal Science Journal*, Vol. 4, 1970, p. 9.

¹² Inal, O. T. and Galligan, J. M., BNL 13820, Brookhaven National Laboratory Reports, Upton, N. Y.

¹³ Damask, A., *Diffusion in BCC Metals*, American Society for Metals, Ohio, 1965, p. 317.