E-10

VARIATIONS IN THE EMBRITTLEMENT OF IRRADIATED PRESSURE VESSEL STEELS

E. T. Wessel[1]

Abstract

The basic aspects of the deformation and fracture characteristics of several heats of ASTM A302B steel were investigated in terms of the Petch parameters. Both irradiation sensitive and insensitive conditions were represented in the data which were analyzed. In the irradiated condition the friction stress, σ_i, increased with increased neutron exposure for all heats of steel. However, the sensitive heats exhibited a much greater increase in σ_i than did the insensitive heats. The grain boundary resistance, k_y, decreased with increased exposure, particularly at the higher exposure levels ($>10^{18}$NVT). The relative changes in σ_i and/or k_y appear to be the predominant factors controlling sensitivity to irradiation damage.

Introduction

The present investigation is part of a broad study of irradiation damage. The immediate objective is to establish which of the basic parameters that control the flow and fracture behavior are responsible for the pronounced variations in sensitivity to irradiation which have been observed.(1) Ultimately, it is expected that it will be possible to control the active parameters through chemistry, heat treatment, grain size, etc. in order to produce heats of steel of the insensitive type only.

Materials and Source of Data

The basic aspects of the deformation and fracture characteristics of several thick plates from various commercial heats of ASTM A302B steel were investigated using the irradiation data which have been generated over a period of several years.(1) The chemical composition of these heats of steel are given in Table I. Other pertinent details concerning the material and irradiation conditions may be found in Reference (1). Without going into details, it may be assured that the observed differences in sensitivity to irradiation embrittlement cannot be traced to possible differences in the conditions employed in the irradiation experiments.(1) The consistent behavior for given heats (103 and 106) in two irradiation experiments may be seen in Fig. 1. In the present investigation an attempt is made to identify and evaluate the influence of the various basic flow and fracture parameters on the increase in yield strength and the associated increase in transition temperature which are observed after the steel is subjected to neutron exposure. Particular attention is given to establishing which of the active parameters are responsible for the marked differences in response to irradiation (sensitive versus insensitive) that have been reported(1) for these different heats of A302B steel (Fig. 1). This present paper consists primarily of a phonomenological description of the influence of the active parameters. It has been possible to identify the major controlling parameters and to associate the "sensitive" and "insensitive" heats with differences in the magnitude of some of these parameters. Subsequent investigations will be directed toward acquiring a more thorough understanding of the factors which affect and control each of these important parameters.

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Experimental Data for Irradiated A320B Steel

(1) The Increase in Yield Strength

In studying the increase of yield strength that occurs upon irradiation (which is also associated with the increase in transition temperature), the following general expression (2-5) is being utilized:

$$\sigma_y = \sigma_i + k_y d^{-1/2} \qquad (1)$$

σ_y = yield strength (#/in.^{2})

σ_i = total friction stress opposing plastic deformation within a grain volume (#/in.^{2})

k_y = force resisting plastic deformation in the region of grain boundaries (#$\sqrt{\text{in.}}$/in.^{2})

d = grain diameter (in.)

First, consideration is given to the effects of irradiation on the total measured yield strength, σ_y. The general behavior of four heats of A302B steel are shown in Fig. 2. All of the tensile properties in this and subsequent figures are from room temperature tests. As seen, the behavior of the heats are similar except that there is a slightly stronger increase in strength in the sensitive heats, particularly at the higher exposure levels. Fig. 3 shows the magnitude of the increase in yield strength ($\sigma_y = \sigma_y$ irrad. - σ_y unirrad.) with increased neutron exposure. The sensitive heats exhibit a significantly greater increase in strength for given exposures, particularly at the higher levels of irradiation. When considered in terms of the percentage increase in yield strength, Fig. 4, there is a very pronounced difference between the sensitive and insensitive heats in terms of their response to irradiation. This increase in yield strength and the difference in response of the various heats is consistent with the observed changes in transition temperature (Fig. 1). A linear correlation exists between the increase in yield strength and the increase in transition temperature as shown in Fig. 5. Of significance is the fact that both the sensitive and insensitive heats lie on the same straight line.

It is now pertinent to relate the yield strength increase to the σ_i and k_y terms of equation (1). Since a systematic study of the effect of grain size in a given heat of steel was not included in the available data (1), it is not possible to use the conventional technique (zero intercept on a σ_y versus $d^{-1/2}$ plot) for determining σ_i. However, it was possible to obtain comparative σ_i values from existing autographic load-extension curves by using an alternate technique. This method is illustrated in Fig. 6. Here σ_i is defined as the intercept between the true stress-true strain curves, determined from the autographic load-extension records, and the elastic behavior. Hull(6) has employed this technique and has found good agreement between σ_i values thus obtained and those derived in the conventional manner. This extrapolation technique has more recently been described by Owens and Hull(7).

The effect of irradiation on the friction stress, σ_i, for four heats of A302B steel is shown in Fig. 7. The change in σ_i, upon irradiation, is shown in Fig. 8. The sensitive heats exhibit a larger increase in σ_i than do the insensitive heats. This is more emphatically demonstrated in Fig. 9 in terms of the percentage increase in σ_i.

The increase in friction stress with increased irradiation is commonly observed and is a generally accepted behavior pattern. Hull and Mogford(8) have observed an increase in σ_i, which approximately corresponded to the increase in yield strength, σ_y, for En 2 steel. A similar behavior was observed for silicon iron by Hull(9). Molybdenum has been studied by Johnson(10) and Wronski and Johnson(11), and it too exhibits an increase in σ_i. Similar increases in σ_i have been reported for pure iron by Campbell and Harding(12) and Mogford and Hull(13).

The rise in σ_i with increased neutron exposure is indicative of an increased resistance to plastic flow within a grain volume. This is attributed to an increase in the number or effectiveness of obstacles which impede the movement of dislocations; such as, precipitates, foreign atoms, point defects, other dislocations and the inherent resistance of the crystal lattice itself. At the present time, it is not possible to define the specific contribution of each of these factors in raising σ_i in the A302B steels. However, in the literature there is fairly good general agreement that a large portion of the increase in σ_i may be attributed to the interaction of moving dislocations with the stress fields around point defects, or clusters of defects created by the irradiation.

Let us now consider the other term, k_y, in the expression (equation 1) for the yield strength. This term depicts the resistance to deformation in the regions of grain boundaries. When obtained in the conventional manner(2-5), k_y is the slope of the yield strength versus $d^{-1/2}$ curve. However, in this case where no systematic data exist, k_y is calculated using equation (1) and the known values of σ_y, σ_i and d. Fig. 10 illustrates the effect of irradiation on the k_y values, obtained in this manner, of four heats of A302B steel. Note that the sensitive heats exhibit a larger initial k_y value and a greater decrease in k_y at the higher exposure levels. The change in k_y is shown in Figs. 11 and 12. Here again there is a distinction between the sensitive and insensitive heats; the sensitive heats exhibiting a gradual decline at exposures up to about 10^{19} NVT and then a rapid decrease at higher exposure levels. On the other hand, the insensitive heats maintain a relatively constant value of k_y up to about 10^{19} NVT, and do not decrease as rapidly at higher exposures.

The decrease in k_y with increased irradiation has not been generally observed, whereas all of the investigators have consistently found σ_i to increase with irradiation. The general conclusion has been that irradiation has little or no effect on k_y. However, in agreement with the present findings, Campbell and Harding(12) have reported a decrease in k_y for irradiated pure iron. A similar behavior is also indicated by some data on molybdenum(10,11,13).

On the basis of current concepts of irradiation damage and fracture, an increase in σ_i would be expected; whereas a decrease in k_y would not be anticipated. For example, in Cottrell's fracture concept (14) involving σ_i and k_y, the transition temperature should be very sensitive to changes in k_y. A large decrease in k_y, such as occurs in the A302B steel, would be expected to substantially lower the transition temperature. The present data would indicate that the opposite occurs. However, it seems entirely possible that irradiation could lower k_y and at the same time raise the transition temperature. The observed change in transition temperature could be the result of the competitive processes of increasing σ_i tending to raise transition temperature while decreasing k_y is tending to lower it. Since the end effect is to raise transition temperature, σ_i must be the predominating factor. The fact that σ_i rises at a faster rate than the total measured yield strength, Fig. 13, would support this contention. It is also possible that another interpretation could be applied to k_y; that is, instead of k_y depicting the resistance to plastic flow at the grain boundaries, it may be indicative of the resistance to crack propagation across grain boundaries. In this case, a decrease in k_y would be expected to raise the transition temperature. Also, it is quite likely that the k_y which is measured may be broken down into several terms, as has been done (2-5) for σ_i. Several other speculative reasons could be advanced, but these are more properly the subject of extended investigations.

(2) The Increase in Transition Temperature

The data relative to measured changes in transition temperature (30 foot pound criterion) are provided in the following figures. Fig. 14 summarizes the available data on the effects of irradiation upon changes in transition temperature for several heats of A302B steel.(1) The relationship of the change in yield strength to the change in transition temperature was already shown in Fig. 5. The increase in friction stress, σ_i, similarly may be related to the change in transition temperature as shown in Fig. 15. Of significance is the fact that both the sensitive and insensitive heats lie on the same straight line. This suggests that σ_i is the predominant controlling factor and that the difference between the sensitive and insensitive heats results from their respective responses of σ_i to irradiation. The change in k_y may be related to the change in transition temperature as shown in Fig. 16. As previously discussed, the reasons for an increased transition temperature with decreased k_y are, at the present, only speculative. The relative behavior of the yield and friction stresses are shown in Fig. 17. Here it is seen that in order to provide an equivalent change in transition temperature, the friction stress must increase to a greater degree than the yield strength. This is consistent with the concept that σ_i must overcome the counteracting effect of a decreasing k_y.

In considering these correlations of the various parameters with the change in transition temperature, it must be emphasized that the change in transition temperature that is shown is the total measured change in transition temperature, which undoubtedly is the composite result of the effect of several factors. Hence, the behavior shown can only be indicative of the general trend, as they do not reflect the individual contribution of the specific parameter being considered. In future investigations it may be possible to obtain a more comprehensive understanding of the role of the individual parameters. Some serious consideration should also be given to the probably influence of irradiation on γ, the effective surface energy for crack propagation. In existing expressions (14,15) relating the change in yield strength upon irradiation to changes in transition temperature, γ is assumed to be unaffected by irradiation. However, in unirradiated steels, the general trend is that an increase in yield strength lowers the effective surface energy for brittle fracture.(16) It seems reasonable that irradiation and the associated increase in yield strength should have a similar effect.

Experimental Data on Unirradiated Steels

Table II lists the pertinent data for the unirradiated, control tensile tests, the samples for which were taken from the same plates of steel and tested at room temperature.

In considering these data with regards to their unirradiated properties, it is apparent that heats 103 and 101 (which are sensitive to irradiation) exhibit lower initial yield strengths, lower friction stress (σ_i), higher k_y's, and higher transition temperatures than do the other (insensitive) heats. Because of their lower initial yield strengths and friction stresses, it is not unreasonable to expect that these parameters might increase more rapidly with irradiation than would those with higher initial values. This would lead to a larger increase in transition temperature when irradiated. Their higher initial k_y (grain boundary resistance) would be expected to result in higher initial transition temperatures, as is apparent from the measured values shown in Table II.

Thus it appears that it may be possible to distinguish which behavior, sensitive or insensitive, will occur upon irradiation from a knowledge of the unirradiated tensile properties and, perhaps, transition temperature. A low initial σ_i when combined with a high k_y (and associated high transition temperature) would be expected to lead to a sensitive type behavior. A moderately high σ_i combined with a low k_y (and low transition temperature) would be expected to be relatively insensitive. Because of the limited amount of data available as a basis for this discussion, it must be emphasized that the preceding suggestions concerning the ability to predict sensitivity can only be tentative. It is anticipated that a more thorough understanding of the influence of the active parameters, i.e., σ_y, k_y, σ_i, and γ, will be of value in establishing the capabilities of predicting and controlling sensitivity to irradiation.

Effects of Chemical and Metallurgical Variables on Basic Flow and Fracture Characteristics

At the present time the work has not progressed to a sufficient extent to be able to isolate and evaluate the specific influence of the innumerable detailed chemical and metallurgical variables with respect to their respective contributions to the σ_i and k_y responses. However, one gross metallurgical difference, and corresponding difference in behaviors of σ_i and k_y was observed and warrants discussion at this time. There is a significant difference in the microstructures of the sensitive and insensitive heats that were included in this investigation (Fig. 18). These differences stem from gross differences in the initial heat treatment; the coarse structures arise from slow cooling from a high austenitizing temperature whereas the fine structures were obtained by rapid cooling from a much lower austenitizing temperature. These photomicrographs are from the irradiated Charpy impact specimens. The unirradiated microstructures are the same as those shown for the irradiated condition. Such large differences in microstructure could not have resulted from irradiation.

The coarse microstructures of the sensitive heats would inherently be less fracture resistant than the fine structures in either the unirradiated or irradiated conditions. In fact, some quantitative measurements of the plane strain fracture toughness, G_{Ic}, had been previously made[2] on heats 106 and 103 (unirradiated) using disk bursting and pre-cracked plate tensile tests. This work demonstrated that the coarse-grained plate of 103 has much less fracture toughness than heat 106; their respective values being 50 and 200 in.lbs/in.^{2}. While the coarse condition is generally undesirable and is

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[2] Unpublished data, G. O. Sankey and E. T. Wessel, Westinghouse Research Laboratories.

not representative of properly processed plate, the data do have some practical significance since this type of microstructure could possibly be representative of the weld areas in actual structures.

Summary

The basic aspects of the deformation and fracture characteristics of several heats of ASTM A302B steel have been investigated using raw data provided by Bettis Atomic Power Laboratory.(1) Both the irradiation sensitive and insensitive conditions are represented in these data.

The classical expression

$$\sigma_y = \sigma_i + k_y d^{-1/2}$$

was employed in an attempt to obtain quantitative information regarding the σ_i and k_y values of the various lots of the same steel which exhibited marked differences in response to irradiation. Since these factors are directly involved in determining flow and fracture characteristics of bcc metals, it was believed that it would be significant to know whether or not the various heats of steel exhibited marked differences in these values, particularly in response to irradiation.

These values have now been determined and some significant differences have been observed. These may be summarized as follows:

1. In the unirradiated condition the sensitive heats exhibit significantly lower yield strength; lower friction stresses, σ_i; higher grain boundary resistance, k_y; and higher transition temperatures than do the insensitive heats.

2. In the irradiated condition

a) The friction stress, σ_i, increases with increased neutron exposure for all heats. However, the sensitive heats exhibit a much greater increase in friction stress, σ_i, than do the insensitive heats, particularly at exposure levels above 10^{18} NVT. This contributes to a larger increase in transition temperature. The increase in σ_i is consistent with the behavior anticipated from current irradiation damage observations and fracture concepts.

b) The grain boundary resistance, k_y, decreases with increased irradiation, particularly at the higher exposure levels ($>10^{18}$ NVT). The sensitive heats have a much higher (~50%) k_y than the insensitive heats except at high exposure levels where k_y for the sensitive heats decreases to values equivalent to those exhibited by the insensitive heats. From existing fracture theory a decrease in k_y normally would be expected to decrease the transition temperature. It is possible that this does occur, but is overridden by a much stronger opposing force - the increase in σ_i. It is also possible that the conventional interpretation of the significance of k_y is not applicable.

3. When these parameters σ_i and k_y are correlated with the change in transition temperature no distinction is made between sensitive and insensitive heats;[3] that is, data from all of the heats lie on the same line. This suggests that the changes in σ_i and/or k_y are the predominant controlling factors in both sensitive and insensitive heats. The individual contributions of σ_i and k_y to the total observed change in transition temperature cannot be determined from the present data. Likewise, the specific factors governing or influencing the total observed behavior of either σ_i or k_y cannot be described at this time.

4. Based on the available data, it appears that the differences in σ_i and k_y and the corresponding bulk differences in irradiation sensitivity may be associated with gross differences in microstructure. The sensitive heats have an abnormal, extremely coarse structure and large grain sizes, whereas the insensitive heats have normal microstructures. The differences in microstructure are attributed to differences in the initial heat treatments given to the various plates of steel.

It is believed that the observed differences in the active parameters between the sensitive and insensitive heats suggest more extensive investigations of the factors which control the magnitude of these parameters; hence the fracture characteristics of the metal. The present work also suggests that some revision in the classical interpretations of the significance of these parameters may be necessary; Armstrong has already published some work (17) in this regard.

Acknowledgements

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9. Hull, D., Acta Met., Vol. 9, p. 413, 1960.
10. Johnson, A. A., Phil. Mag., Vol. 5, p. 413, 1960.

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[3] The distinction between sensitive and insensitive is only apparent in terms of the magnitude of the change of σ_i and/or k_y at some given exposure level. The greater the change in these parameters, the greater the change in transition temperature.

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TABLE I

Chemical Composition (weight percent) ASTM A302 Class B Steel

Heat No.	C*	Mn**	Si	Cr	Mo	Al	Ni	P**	S**	N***	O□
101	0.22	1.31	0.25	0.18	0.28	0.046	0.37	0.014	0.014	0.004	0.0023
102	0.22	1.34	0.31	0.09	0.32	0.032	0.05	0.014	0.022	0.004	0.0016
103	0.26	1.37	0.32	0.10	0.42	0.038	0.21	0.028	0.029	0.005	0.0022
104	0.26	1.36	0.30	0.14	0.31	0.045	0.26	0.015	0.024	0.007	0.0011
105	0.26	1.42	0.28	0.12	0.33	0.028	0.17	0.010	0.024	0.006	0.0016
106	0.21	1.35	0.34	0.21	0.33	0.050	0.16	0.013	0.014	0.010	0.0057

* Combustion method
** Wet analysis
*** Kjeldahl method
□ Vacuum fusion
All others by spectrochemical means

TABLE II

Heat No.	Class	Yield Strength, psi		σ_i (psi)	k_y (psi $\sqrt{\text{in.}}$)	Transition Temp. (30 ft lb) °F
		σ_y .01%	σ_y 0.2%			
106	Insensitive	55,000	52,000	33,750	802	-24
105	Insensitive	69,000	69,000	48,500	1005	30
104	Insensitive	67,000	67,800	53,500	--	-30
103	Sensitive	32,000	42,000	25,200	1515	100
101	Sensitive	37,200	43,600	24,750	1790	88

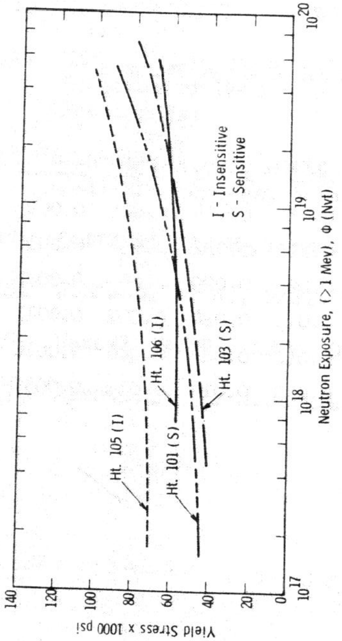


Fig. 2—The effect of irradiation on yield strength

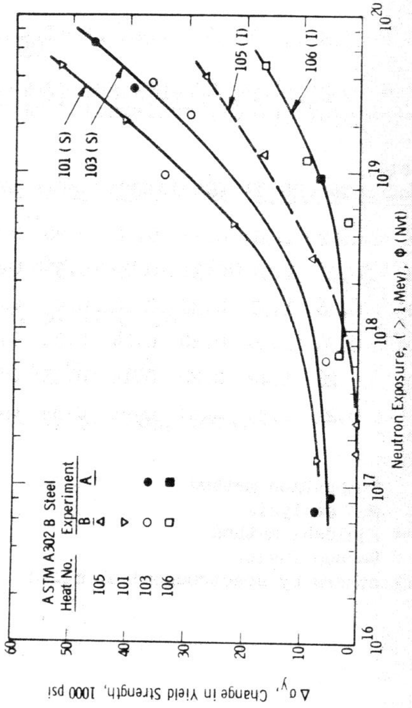


Fig. 3—The effect of irradiation on the change in yield strength

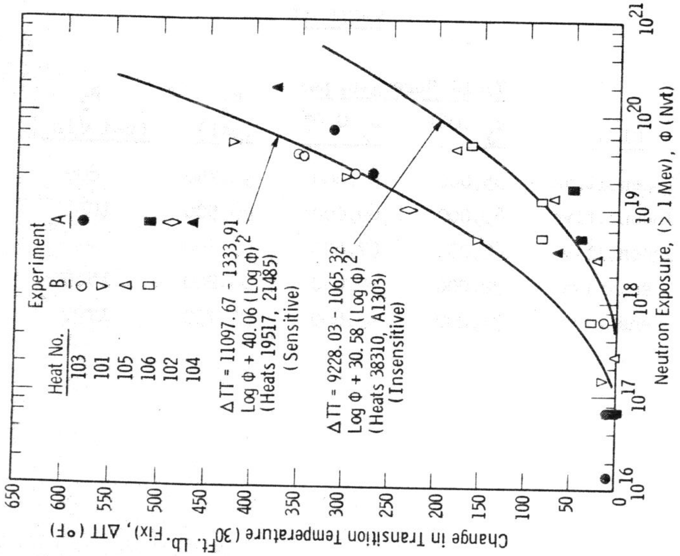


Fig. 1—The effect of irradiation on various heats of A302B steel (Ref. 1)

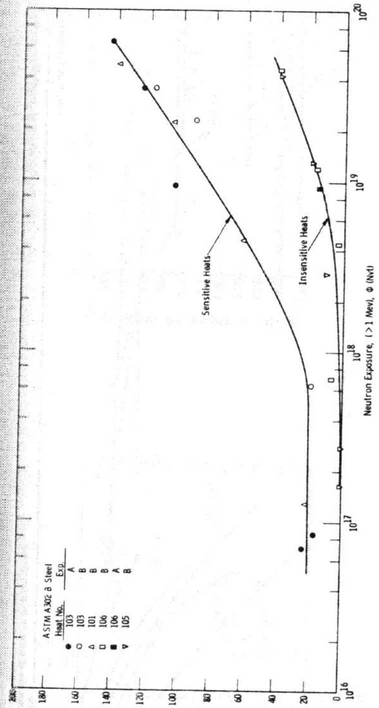


Fig. 4—The effect of irradiation on the percentage change in yield strength

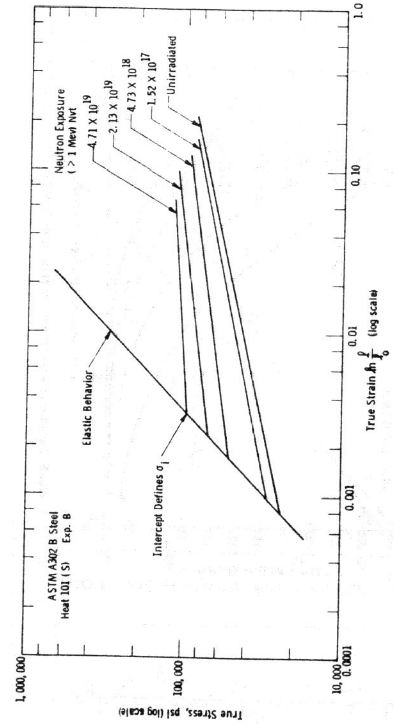


Fig. 6—Determination of σ1 by extrapolation technique

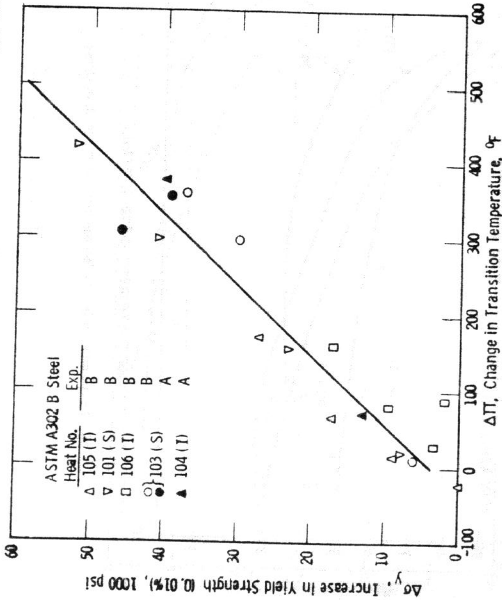


Fig. 5

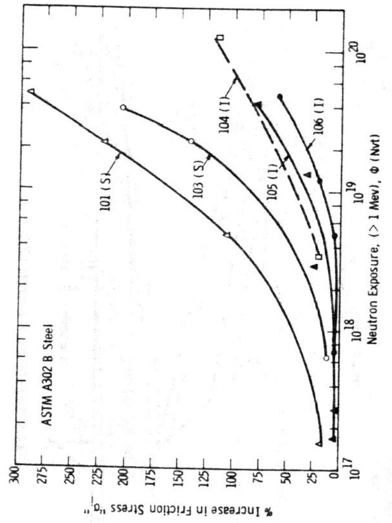


Fig. 9—The effect of irradiation on the percentage change in friction stress

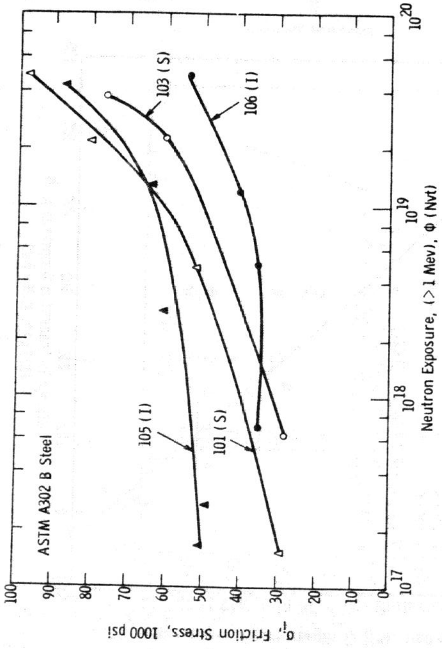


Fig. 7—The effect of irradiation exposure on the friction stress

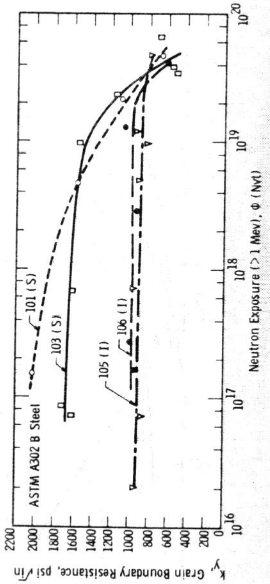


Fig. 10—The effect of irradiation on the grain boundary resistance to yielding

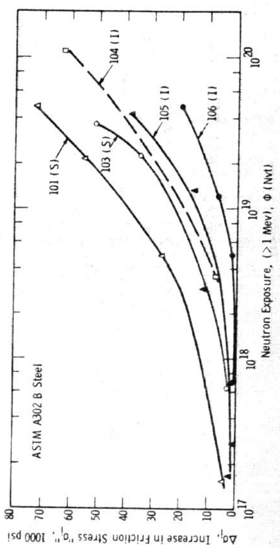


Fig. 8—The effects of irradiation on the change in friction stress

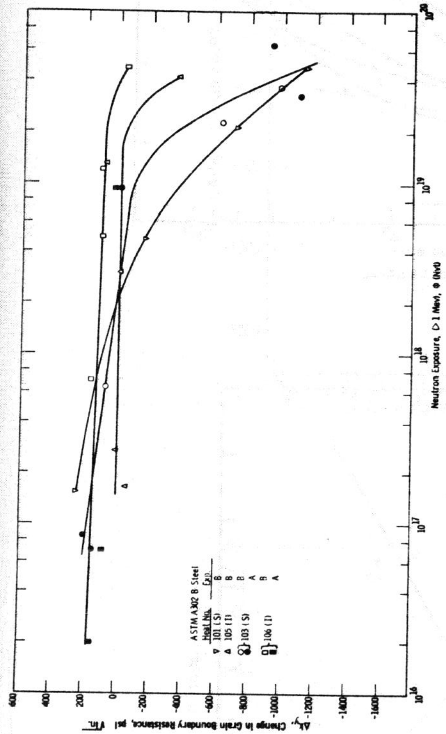


Fig. 11—The effect of irradiation on the change in grain boundary resistance

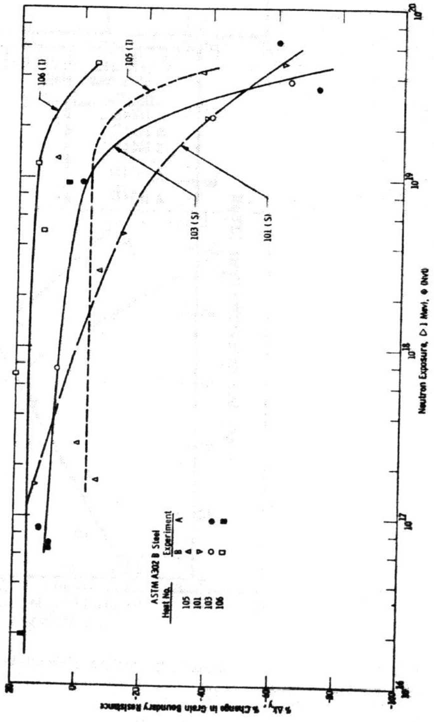


Fig. 12—The effect of irradiation on the percent change in grain boundary resistance

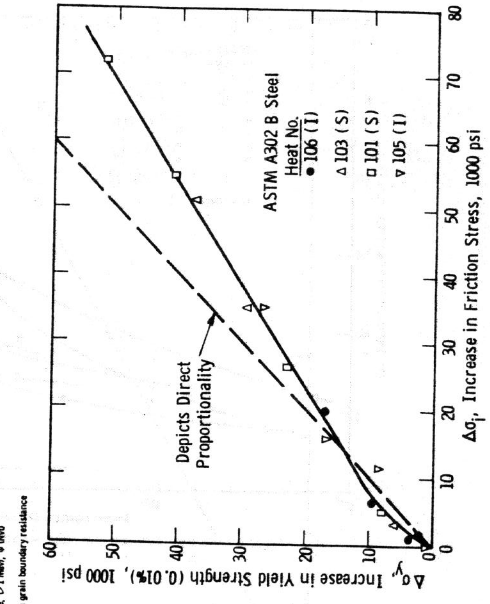


Fig. 13—Relative changes in yield strength and friction stress

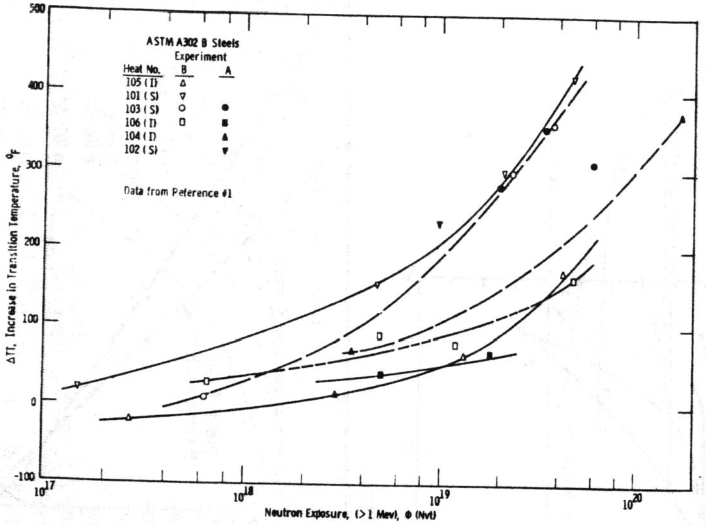


Fig. 14—The effect of irradiation on transition temperature (30 ft. lb. criterion)

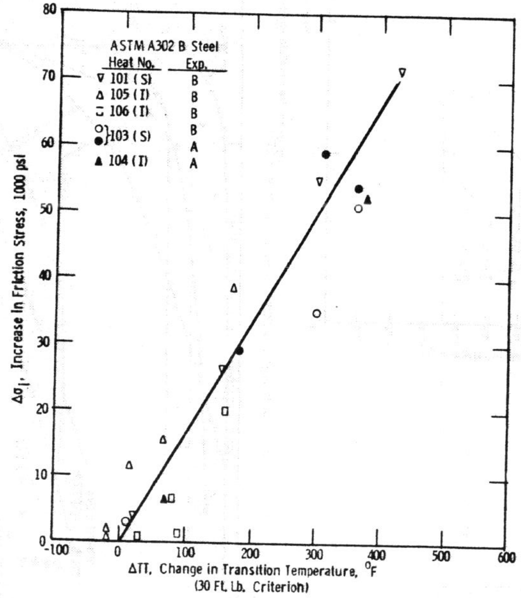


Fig. 15

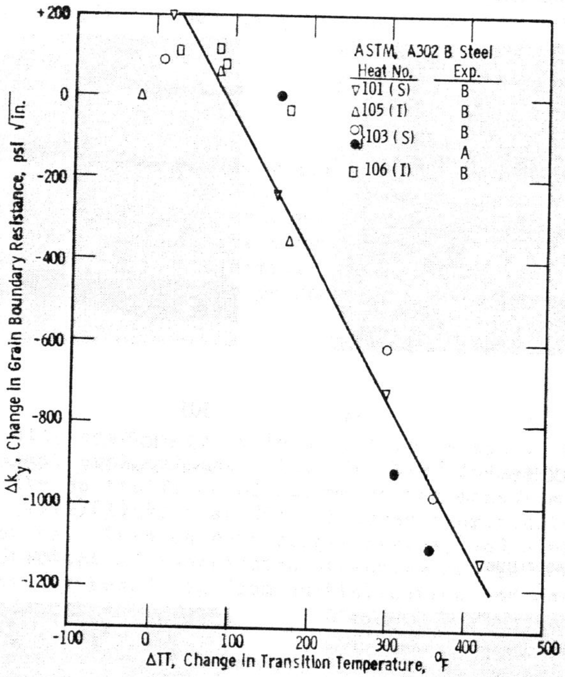


Fig 16

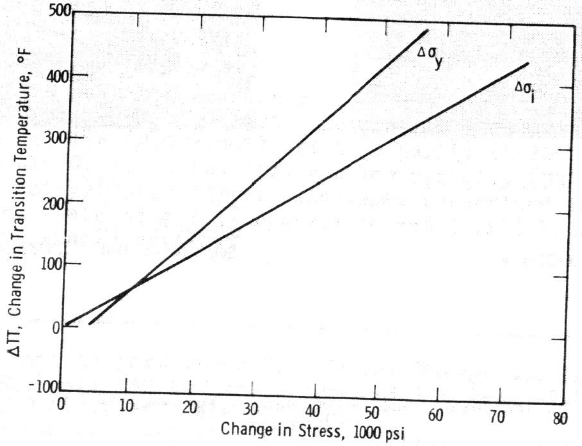
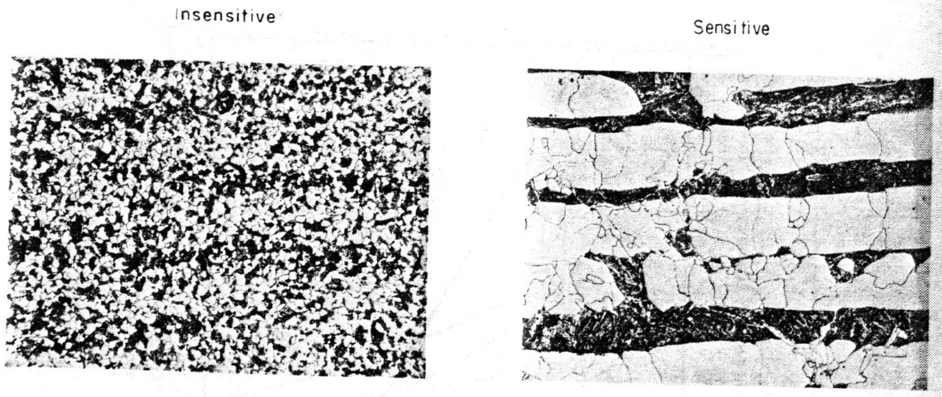


Fig. 17—The relative behavior of the yield and friction stresses



Insensitive

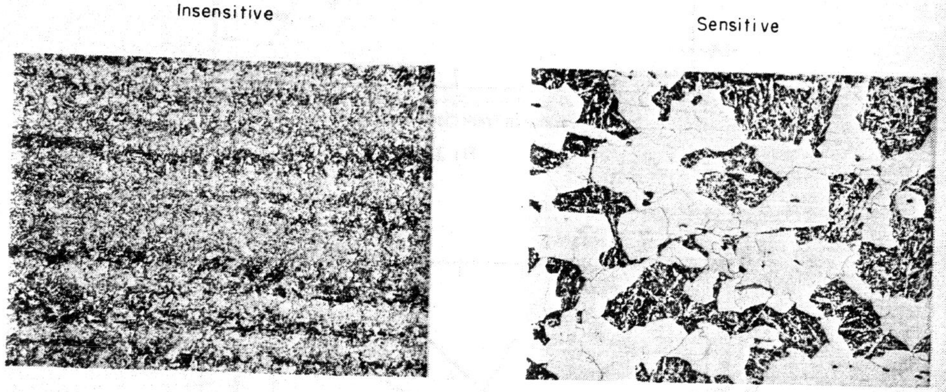
Sensitive

106 84X

4.5×10^{19} Nvt
Avg. Grain Dia. .00166 in.

101 84X

4.5×10^{19} Nvt
Avg. Grain Dia. .0095 in.



Insensitive

Sensitive

105 84X

5.0×10^{19} Nvt
Avg. Grain Dia. .0024 in.

103 84X

5.0×10^{19} Nvt
Avg. Grain Dia. .0075 in.

Comparison of irradiated microstructures of various heats of ASTM A302B steel.

Fig. 18