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NEUTRON EMBRITTLEMENT OF REACTOR PRESSURE VESSEL STEELS

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ABSTRACT

This report presents the status of observations at the U. S. Naval Research Laboratory on the embrittlement of steels which are commonly used for the primary pressure containment vessels of nuclear power plants. The demonstrated criterion of nil ductility transition (NDT) temperature provides the basis for meaningful analysis of neutron-induced embrittlement in reactor steels. Results to date indicate that the degree of embrittlement depends upon the material, the neutron exposure, and the temperature during irradiation. These same variables also affect the degree of notch ductility recovery effected by postirradiation heat treatment. In addition, the time and temperature of heat treatment have been shown to play an important role in establishing the recovery pattern.

The validity of these experimental observations are being tested through correlations with data from reactor surveillance programs and from specimens of the SL-1 reactor pressure vessel. Preliminary data from dosimetry in the SM-1A reactor permit the extension of experimental data to predict the increase in NDT of the reactor pressure vessel.

PROBLEM STATUS

This is a status report on several phases of the problem; work on the problem is continuing.

AUTHORIZATION

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INTRODUCTION

In the first phase of concentrated development of nuclear power reactors, accelerated investigations were undertaken to determine the effects of high energy nuclear particles upon the properties of the major reactor core components, especially fuel and fuel cladding materials. Belatedly, it was observed that significant changes in the properties of structural materials also developed, especially in regions near the core. Accordingly, an accelerated experimental program for the study of neutron irradiation effects on reactor pressure vessel steels has been undertaken.

The marked decrease in the notch ductility of irradiated steels is considered the major deleterious change in the mechanical properties. The significance of these changes is reviewed in reference to the nil ductility transition (NDT) temperature concept. Recent advances in fracture analysis relating stress, flaw size, and temperature conditions for brittle failure of steel structures lends additional engineering significance to irradiation embrittlement data.

The major commercial nuclear power systems in the U. S., pressurized and boiling water reactors, as well as prototypes such as nuclear superheat and high temperature gas-cooled reactors, have in common a ferritic steel pressure vessel. These various nuclear systems represent a wide range of thermal and nuclear exposure conditions which must be considered systematically in order to fully assess the effects of radiation on this critical component.

The several pressure vessel steels which have been studied represent the pressure vessel materials of the majority of current commercial nuclear power plants. The observed embrittlement of these steels is reviewed in summary with emphasis upon the results in terms of materials and conditions of irradiation. Also, postirradiation heat treatment for the restoration of notch ductility has been investigated over a range of irradiation and annealing conditions. The results of these studies are reviewed with emphasis upon specific cases which demonstrate trends as well as certain anomalies. The implications of these data are reinforced by correlation with dosimeters, with specimens from operating nuclear power plants, and with specimens from the SL-1 reactor pressure vessel.

NOTCH DUCTILITY DETERMINATION AND SIGNIFICANCE FOR NUCLEAR APPLICATIONS

During the past fifteen years the problem of the brittle fracture of large steel structures has been studied in considerable detail at NRL by Pellini and his co-workers. This study evolved the nil ductility transition (NDT) temperature determination based upon the drop weight test (ASTM Recommended Practice E-208-63T) (1) and culminated recently in the development of a unified fracture analysis concept (2). The concept relates the key factors of steel fracture: crack initiator flaw size, applied local stress, and service temperature at the time of fracture. The analysis system, which is based upon hundreds of steel structural failures and is referenced to the NDT temperature, has been reduced by Pellini to a generalized fracture analysis diagram applicable to all the steels currently

studied for irradiation embrittlement. This analysis has been extended to demonstrate an application to fracture-safe design involving various degrees of engineering conservatism and to the selection of a steel to meet design objectives (2).

The value of the NDT concept and the related fracture analysis diagram to neutron irradiation embrittlement resides in the direct correlation of data from Charpy V-notch specimens and drop weight test determinations of the NDT. This correlation holds also after irradiation. The irradiation correlation was based upon results obtained by the simultaneous exposure of drop weight test and Charpy V specimens in twenty irradiation experiments with neutron exposures ranging up to 3×19^{19} n/cm^2 (neutron energy >1 Mev). This correlation has been described in detail in earlier publications (3,4).

The major significance of the fracture analysis diagram for nuclear pressure vessels abides in the very rapid rise in stress requirements for fracture of steel structures at temperatures above the NDT. This rise is demonstrated by the fact that, at NDT + 60°F, stresses at or above the yield point of the steel are required for crack propagation. Thus, for the types of steel used in reactor pressure vessels, a point for relative safety from brittle fracture has been established. The question then arises as to how this point may be determined for the pressure vessel of an operating nuclear power plant. The answer to this question has been the primary objective for the study of neutron embrittlement at NRL.

There is no practical way to assure that flaws of significant sizes (inches) are not present in operating reactor pressure vessels. Therefore, the problem of potential brittle fracture must be controlled by minimizing stresses in the vessel while at the same time maximizing the vessel temperature. These objectives are difficult to attain, especially in pressurized water reactors, because of the rapid rise in pressure requirements to keep water from boiling at progressively higher operating temperatures. The best possible knowledge of the NDT temperature increase with neutron irradiation must be applied in conjunction with the knowledge of stress conditions to overcome the lack of knowledge of flaw sizes for minimizing the possibilities for fracture propagation in nuclear pressure vessels. The definition of NDT increases in reactor steels after neutron exposure at various temperatures is the primary purpose of this report.

NEUTRON EMBRITTLEMENT OBSERVATIONS

Experimental Irradiation Techniques

A detailed review of experimental irradiation procedures is beyond the scope of this paper. Nevertheless, some general description of the experimental approach and irradiation techniques is presented in the Appendix as background for the presentation of the neutron embrittlement data. The major experimental factors considered are encapsulation techniques, temperature control and measurement procedures, techniques for determining the nuclear environment, and postirradiation evaluating operations.

As with most programs for the determination of nuclear radiation effects on materials, the experimental time scale is condensed relative to the engineering application condition such that most of the experiments involve high irradiation rates for relatively short duration, whereas the components of an actual reactor may receive an equivalent dosage over a period of years. This factor will be discussed in more detail in a later section dealing with the application of experimental data to the reactor operating condition.

In every phase of the study, the reliability of the data has been tested by the use of a large number of test specimens and, wherever possible, through repeat experiments under similar thermal and nuclear conditions. Furthermore, control tests have been conducted outside the reactor to assure that thermal effects are isolated from nuclear irradiation

effects. Experimental nuclear environments have been characterized as well as possible within the state of the art through the use of activation type neutron dosimeters in all experiments.

Materials Studied

A continuous effort has been exerted to obtain steels in several forms, that is, as welds, forgings, and plate, which simulate as nearly as possible the materials actually used in the construction of nuclear pressure vessels. In most cases the steels studied are only representative of the types used in pressure vessels, but several segments have been obtained from test sections of heats used in reactor construction. The overwhelming preponderance of power reactor pressure vessels constructed of the ASTM Type A212-B and A302-B steels is demonstrated by the listing in Table 1 (5). This list represents essentially all the commercial plants currently operating or in advanced design or construction stages which have ferritic steel vessels. For comparison, however, other steels of potential interest for nuclear applications have also been investigated.

In the NRL study, emphasis has been placed on the most commonly used steels, ASTM A212-B and A302-B, and on the Army reactor steels, A350-LF1 (Mod.) and A350-LF3. A description of these steels which have been studied comprehensively is provided in Table 2. Steels which have received lesser emphasis are listed in Table 3. In addition, specimens of the A212-B steel from the pressure vessel of the Army SL-1 reactor have been evaluated. Higher strength steels are now being obtained for future study as potential materials for structural application in advanced nuclear power reactors.

Table 1
Power Reactor Pressure Vessel Steels
(Compiled from tabulations of DiNunno and Holt (5))

Pressure Vessel Steel	Reactor	Thickness of Vessel (in.)
A212-B	Vallecitos ESR	3.5
A212-B	Pathfinder	3
A212-B	Piqua	1-1/8 (in region of max flux)
A212-B	Vallecitos BWR	3-3/8
A212-B	Consolidated Edison	6.94
A212-B	BONUS	3-3/8
A212-B	Saxton	5 (Multilayered vessel)
A212-B	SM-1	2.5 (1/4 in. of cladding)
A302-B	Humboldt	4 - 5
A302-B	Dresden	5.375
A302-B	Big Rock Point	5.25 (Exclusive of cladding)
A302-B	Elk River	3
A302-B	Yankee	7-7/8
A302-B	PWR	8-3/8 (1/4 in. of cladding)
A350-LF1 (Modified)	SM-1A	2-1/2 (1/8 in. min and 1/4 in. max cladding)
A350-LF3	PM-2A	2-3/8 (3/16 in. min and 1/4 in. max cladding)

Table 2
Description of Major Steels in Irradiation Study

Item	Steel	Form	Thickness (in.)	Chemical Analysis (%)								
				C	Mn	Si	P	S	Ni	Cr	Mo	Other
1	A302-B	Plate	6	0.20	1.31	0.25	0.012	0.023	0.20	0.17	0.47	-
Heat treatment: Austenitized at 1650° F for 2 hours; water quenched; tempered at 1200° F for 6 hours; furnace cooled to below 600° F												
2	A212-B	Plate	4	0.26	0.76	0.24	0.011	0.031	0.22	0.20	0.02	-
Heat treatment: Austenitized at 1650° F for 2 hours; water quenched; tempered at 1175° F for 4 hours; furnace cooled to below 600° F												
3	T-1	Plate	2	0.13	0.85	0.24	0.013	0.013	0.64	0.67	0.40	0.06 V
Heat treatment: Austenitized at 1700° F for 2 hours; water quenched; tempered at 1150° F for 2 hours; re-tempered at 1165° F for 2 hours; air cooled												
4	HY-80 (Ni-Cr-Mo)	Plate	3	0.14	0.21	0.19	0.011	0.014	2.91	1.55	0.54	0.04 V 0.06 Al
Heat treatment: Austenitized at 1650° F for 3 hours; water quenched; tempered at 1175° F for 3 hours; air cooled												
5	A350-LF1 (Modified)	Plate	3-5/8	0.15	0.79	0.25	0.027	0.033	1.71	0.05	0.04	0.088 Cu. 0.04 V
Heat treatment: Forged in the temperature range of 1700° F min to 2250° F max. Post-forging treatment: normalized at 1600° F; water quenched; tempered at 1250° F; stress relieved at 1150° F												
6	A350-LF3	Forging	2.4	0.14	0.52	0.25	0.031	0.032	3.28	0.04	0.05	0.04 V
Heat treatment: Forged in the temperature range of 1700° F min to 2250° F max. Post-forging treatment: normalized at 1550° F; water quenched; tempered at 1200° F; stress relieved at 1150° F												

Table 3
Additional Steels Investigated for Neutron Embrittlement

Item	Steel	Form	Thickness (in.)
1	A302-B Base Plate	Submerged Arc Weld	4
2	A302-B Base Plate	Submerged Arc Weld	5-1/2
3	A212-B Base Plate	Submerged Arc Weld	6-1/2
4	A212-B	Plate	6-1/2
5	A201	Plate	2
6	A201	Plate	3
7	A201	Plate	6
8	A336	Forging	4
9	A353	Plate	1
10	17-4 PH (H-1100)	Rod	5/8

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Experimental Observations Related to Materials,
Neutron Exposure, and Irradiation Temperature

The materials engineer who recommends a steel for a reactor pressure vessel, among many other considerations, must review the problem of the NDT temperature. His first consideration in this connection involves the determination, as nearly as possible, of the as-fabricated NDT temperature. He must then add to this determination the best judgment as to the increase in NDT temperature (and the attendant operating limitations) which can be expected for the chosen steel under nuclear and thermal conditions for the specific reactor pressure vessel. To date, however, the factors of proven experience plus structural strength and fabricability in heavy sections have effectively limited the selection of commercial reactor pressure vessel steels. Table 1 demonstrated the heavy dependence upon the two steels, A212-B and A302-B. With more comprehensive data on irradiation effects in these and other pressure vessel candidate steels, the factor of nuclear radiation damage may play a bigger role in the selection of reactor structural components. Accordingly, data on neutron embrittlement of selected steels is presented to demonstrate the differences in effects which can be related to the material, the neutron exposure, and the temperature during irradiation.

Materials – Initial experiments involved the irradiation of several steels at relatively low temperatures (<450° F) with the objective of demonstrating whether or not significant data trends could be observed. A general and consistent relationship between embrittlement and neutron exposure became apparent. This relationship is demonstrated for several different steels in Fig. 1. The data pattern at first appears to be independent of the steel type. However, as the experimental pattern of irradiation at low temperature to several different neutron dosages was applied to each type of steel, data trends related to differences in the steels were indicated. The NDT data presented in Fig. 1 have been separated by materials and are presented along with elevated temperature data in Figs. 2-6. The shaded scatter band in each figure represents the general trend for NDT temperature increase for low temperature irradiations (<450° F) and with neutron exposure in terms of n/cm^2 (neutron energy >1 Mev). Data on the A201 steel is not summarized separately. In these five figures the data points are identified with a number which represents the irradiation temperature. Solid points were obtained with instrumented (controlled and measured temperature) irradiation assemblies, while the open points represent capsule irradiations in which the temperature was estimated on the basis of low melting thermal monitors.

The difference in embrittlement is particularly apparent when all data points for the A350 steels (Fig. 2) are compared with data for the T-1 and HY-80 steels (Fig. 3). The trend line for the embrittlement of the A350 steels approximates very closely the upper boundary of the general trend band, while the lower boundary represents the trend for embrittlement of the quenched and tempered T-1 and HY-80 steels. The summary for low temperature irradiation of the A212-B (Fig. 4) and A302-B (Fig. 5) steels, as well as for the weld metal used in joining plates of these steels (Fig. 6), demonstrates an intermediate embrittlement behavior. The data for the A302-B indicate a somewhat greater sensitivity to embrittlement than does the A212-B. However, it should be noted that most of the data for the A302-B were obtained on one particular heat whereas the A212-B data involve two plates of different thickness from different heats. Even with the A212-B, the data are quite limited for comparison among different heats of the same nominal type. Nevertheless, with the wide range of materials studies (Fig. 1) and the consistent trends observed, it is difficult to postulate major differences in irradiation embrittlement among heats of the same nominal composition.

One experiment involving the HY-80 steel required a departure from the normal mode of determining the NDT temperature from the Charpy curve. For the 3-inch HY-80 plate studied, the NDT point is at the 70 ft-lb level on the Charpy curve. However, after irradiation to approximately 8×10^{19} n/cm^2 (>1 Mev) at about 250° F, the ductile portion of the Charpy curve dropped to the 40 ft-lb level (Fig. 7). In this case, a shift in the NDT of 490° F

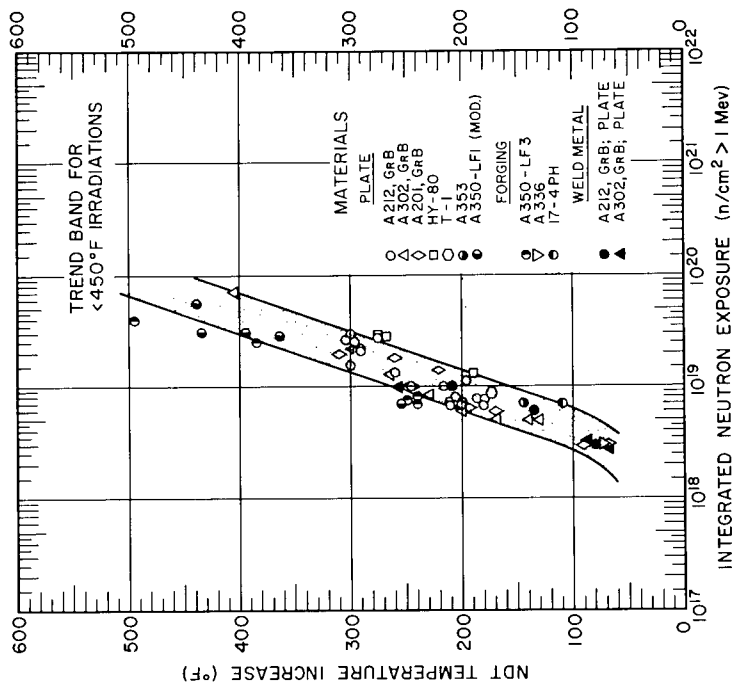

Fig. 1 – Increase in the NDT temperatures of steels resulting from irradiation at temperatures below 450°F

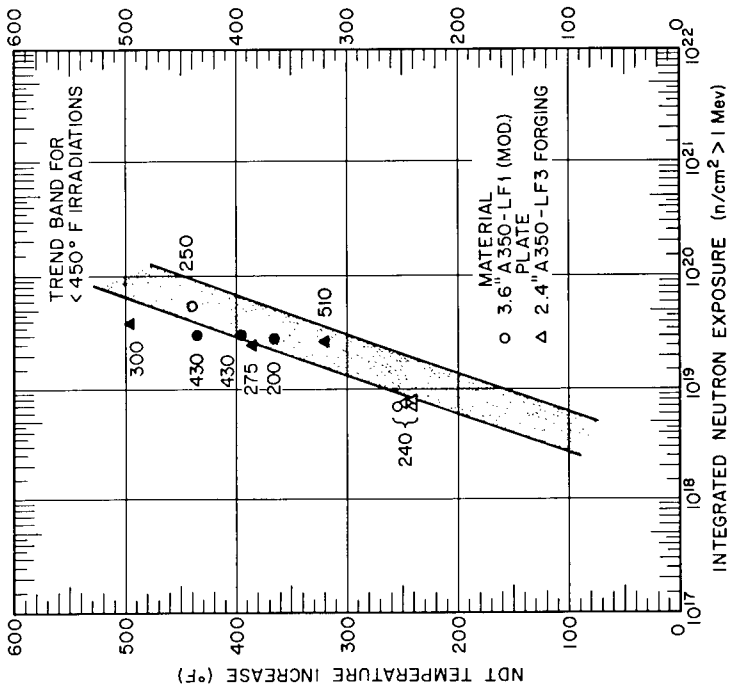

Fig. 2 – Increase in the NDT temperatures of A350 steels resulting from neutron irradiation at various temperatures

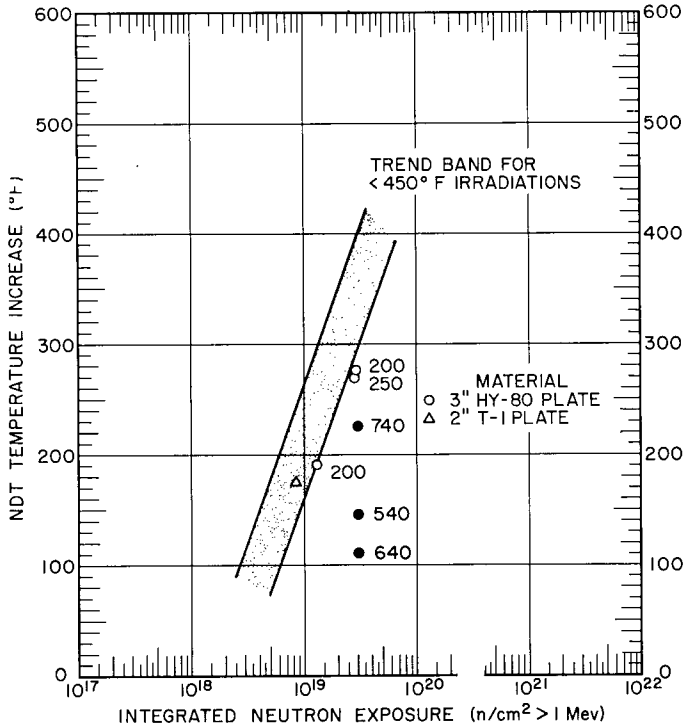

Fig. 3 – Increase in the NDT temperatures of quenched and tempered (HY-80, T-1) steels resulting from neutron irradiation at various temperatures

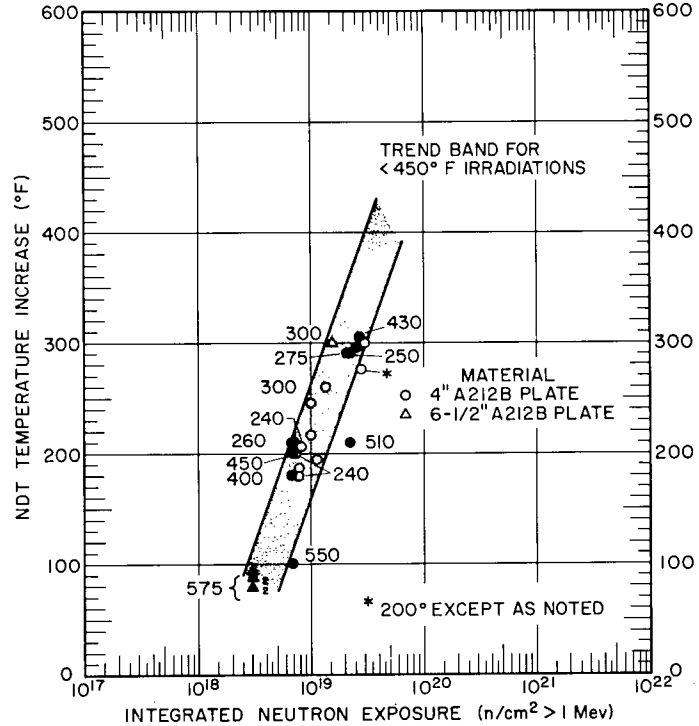

Fig. 4 – Increase in the NDT temperatures of A212-B steels resulting from neutron irradiation at various temperatures

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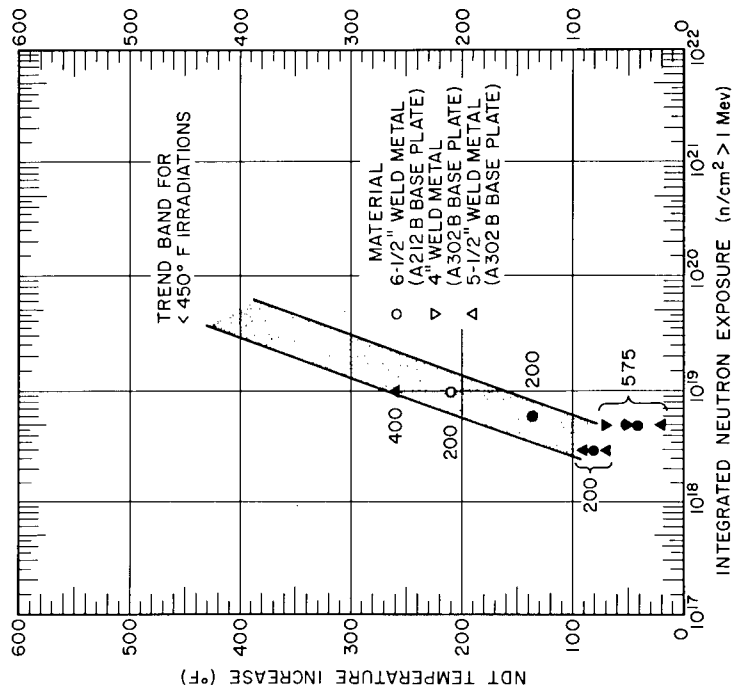

Fig. 6 – Increase in the NDT temperatures of weld metals resulting from neutron irradiation at various temperatures

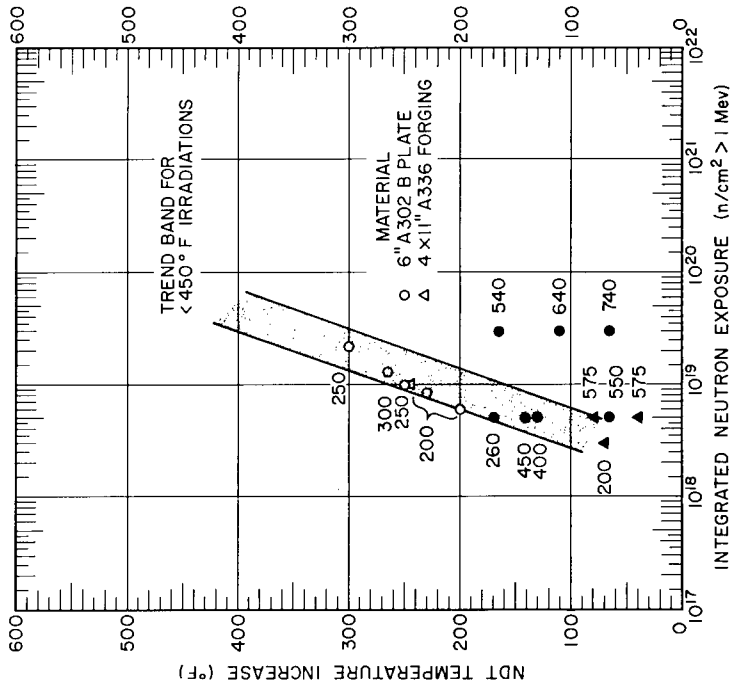

Fig. 5 – Increase in the NDT temperatures of A302-B and A-336 steels resulting from neutron irradiation at various temperatures

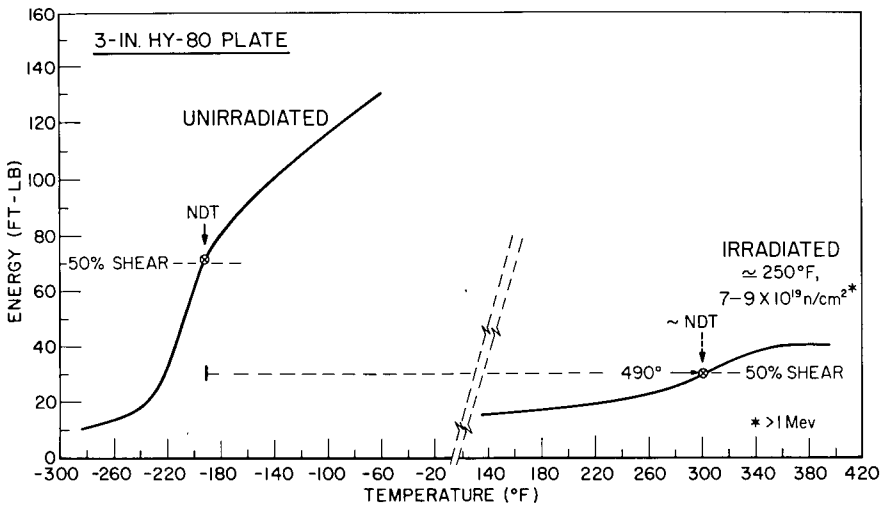

Fig. 7 – Effects of high neutron exposure on transition temperature and shear energy characteristics of HY-80 (Ni-Cr-Mo) steel. (Neutron exposure estimated from prior irradiation experiments = 7 to 9 $\times 10^{19}$ n/cm^2 > 1 Mev.)

was estimated from the respective 50-percent shear points on the before and after irradiation Charpy curves. The drop in the ductile shelf of the Charpy curve is characteristic of irradiated steels, but the extreme is observed with the HY-80 irradiated under these conditions. This observation introduces another factor in the analysis of neutron embrittlement. In addition to the ΔNDT after irradiation, the reduction in the full shear energy portion of the Charpy curve must be considered because of the possibility of low energy shear fracture.

Differences in the irradiation response of several steels have been tested through a capsule irradiation experiment. Five steels, A212-B, A350-LF1(Mod.), A350-LF3, A353, and 17-4 PH, with compositions which differ primarily in nickel and chromium content (Table 4) were irradiated simultaneously at 240° F in the Low Intensity Test Reactor (LITR) to a neutron exposure of 7 $\times 10^{18}$ n/cm^2 (>1 Mev). The NDT temperature shifts observed are listed in Table 5. The results of this experiment reinforce the observation of greater NDT increases for the A350 steels and, at the same time, show a considerably smaller change for the A353 and the 17-4 PH steels in comparison with the A212-B reference steel. Results by Trudeau (6) on various ferrites have indicated a similar pattern with greater neutron induced changes in a 3.2 percent nickel ferrite. With higher nickel content, however, a reverse trend to lower embrittlement is observed. It is apparent that further study is required to determine whether or not the observed results are simply composition differences or, as is more likely, are combined effects of microstructure and composition.

The differences in the degree of embrittlement observed with the several steels investigated gives rise to the encouraging possibility of finding a pressure vessel steel which is less sensitive to radiation than those currently in use. It should be noted, however, that within the limits of available data, much greater advantages are to be gained by finding a satisfactory pressure vessel steel which has a very low initial NDT temperature. For example, if a good pressure vessel steel could be found with an initial NDT of -200° F, the question of differences in sensitivity to neutron embrittlement of the degree noted would be purely academic for most power reactors. The ideal situation would be low initial NDT as well as low sensitivity to irradiation embrittlement. This goal is being pursued in a planned program for the investigation of radiation damage to higher strength steels.

Table 4
Composition of Five Irradiated Steels

Type of Steel	Composition (wt -%)							
	C	Mn	Si	Ni	Cr	Mo	P	S
A212-B	0.26	0.76	0.24	0.22	0.20	0.02	0.011	0.031
A350-LF1 (Mod.)	0.15	0.79	0.25	1.71	0.05	0.04	0.027	0.033
A350-LF3	0.14	0.52	0.25	3.28	0.04	0.05	0.031	0.032
A353	0.10	0.47	0.29	8.99	0.06	0.15	0.008	0.023
17-4 PH	0.03	0.30	0.60	4.24	16.36	-	0.019	0.015

Table 5
Increase in NDT for Five Irradiated Steels
(Irradiated at 240° F to a neutron dosage of 7×10^{18} n/cm^2 (>1 Mev).)

Material	ΔNDT (°F)
A212-B	200
A350-LF1 (Modified)	255
A350-LF3	240
A353	145
17-4 PH	110

Neutron Exposure – The most elusive variable in the study of radiation effects on materials is the neutron environment, and the importance of this factor can hardly be overstated. In spite of past difficulties, however, developments in threshold or activation type neutron dosimetry in recent years have been very encouraging.

The major questions regarding the nuclear environment are: (a) what criterion for reporting neutron exposure most realistically describes the effective or damaging dosage, and (b) do the nuclear conditions measured in research or test reactor experiments accurately represent exposure conditions in critical components of operating power reactors. These questions are not independent and may be stated alternately as questions of whether there are significant effects of different neutron spectra and of different rates of irradiation. The question of what time-integrated neutron dosage properly represents the anticipated power reactor pressure vessel exposure is also a part of the second question. Table 6 presents peak dosages expected in the power reactors listed in Table 1. The results presented in Table 6 were provided by a survey of the U. S. Atomic Energy Commission in 1961 (5). The anticipated peak pressure vessel neutron exposure for the sixteen commercial and military reactors listed ranges between 5.6×10^{17} and 2.13×10^{20} n/cm^2 (>1 Mev). Of these sixteen reactors only seven have indicated lifetime exposures greater than 1×10^{19} n/cm^2 (>1 Mev). Nevertheless, the exposures expected are sufficient to suggest

Table 6
Neutron Exposure for 16 Power Reactors Having Ferritic Steel Pressure Vessels (5)

Reactor	Estimated Integrated Neutron Dose Expected for Vessel Life n/cm^2 (>1 Mev)	Method of Establishing Neutron Exposure
Vallecitos ESR	5.6×10^{17} (belt line)	Calculation
Pathfinder	1.14×10^{18} (belt line)	Calculation
Piqua	3.4×10^{18}	Calculation
Vallecitos BWR	5.4×10^{18} (belt line)	Measurement at flux window
Consolidated Edison	7.5×10^{18} (belt line)	Calculation
BONUS	9.13×10^{18} (belt line)	Calculation
Saxton	9.5×10^{18} (belt line)	Calculation
SM-1	1.09×10^{19} (for 20 yr at load factor, belt line)	Calculated values normalized to mockup measurements
Humboldt	1.0×10^{17} (belt line)	Calculation
Dresden	9.2×10^{17} (belt line)	Calculation and measurement
Big Rock Point	1.24×10^{19} (belt line)	Calculation
Elk River	1.9×10^{19} (belt line)	Calculation
Yankee	2.04×10^{19} (belt line)	Calculation
PWR	6.0×10^{19} (core 1)	Calculation and measurement
SM-1A	8.05×10^{19} (for 20 yrs at load factor, belt line)	Calculated value normalized to SM-1 mockup measurements, with 30% addition
PM-2A	2.13×10^{20} (for 20 yrs at load factor, belt line)	Calculated value normalized to SM-1 mockup experiments

that direct measurements should be conducted to supplement flux calculations and further, that the extent of radiation damage which may be anticipated in some reactors is significant in terms of the question of operational integrity of the reactor vessel in the planned lifetime.

Much has been written on the subject of measuring the nuclear environment and on relating measurements to radiation damage, but straightforward answers to these key questions are not readily found. Nevertheless, with the steadily growing store of data on this problem, answers to these and other questions will be forthcoming. Even now certain conclusions are possible regarding the neutron exposure as a key factor in assessing radiation damage to metals and alloys.

The commonly accepted answer to the question of which criterion to use in reporting neutron exposure is that of reporting integrated neutron exposure in terms of the dosage of neutrons per square centimeter having energies of one million electron volts or greater (n/cm^2 >1 Mev) as in Table 6. In the NRL study, the 1 - Mev value is determined by the extrapolation of data from threshold or activation dosimeters, as described in the Appendix. This study has involved experiments in eleven different irradiation facilities of five test reactors as well as surveillance irradiations in two Army power reactors. While most of these studies involved close proximity to fuel in a light water medium, the quite different neutron spectrum and irradiation rate conditions of the air-cooled, graphite-moderated Brookhaven reactor were also used. For these experiments, the increase in NDT has been related to the 1 - Mev neutron dosage with the resultant data trends of Figs. 1-6. Only in the case of one irradiation in the Brookhaven Graphite Reactor (BGR) at 280°F did the data depart from the established trend band. The results of this experiment are presented in Fig. 8 with the BGR data plotted along with the shaded trend band of Fig. 1. It is noted that NDT shifts for the highly thermalized spectrum (BGR), thermal to fast (T/F) ratio of 40/1, falls very close to the top edge of the band established for the LITR and MTR (Materials Testing Reactor) irradiations which have a thermal to fast ratio of approximately 2.5/1. In addition to the large differences in neutron energy spectrum, the experiment also included 10/1 difference in dose rate, i.e., the BGR experiment required an exposure approximately ten times that of the experiments related to the band. These results indicate that dose rate and spectrum effects are not significant for one order of magnitude differences in reactor operating conditions. It is believed that this order of difference adequately represents spectrum differences that can be expected for operating power reactors. Investigations to two orders of magnitude differences in dose rate would, in effect, require exposure of steel specimens in power reactors.

In the interest of demonstrating the effect of time-integrated neutron dosage to NDT increases, the data for the A212-B and A302-B plate which were presented in Figs. 4 and 5 are plotted on a linear scale in Fig. 9. The marked tendency toward smaller NDT increases for each additional increment of neutron dose at temperatures below 450°F is readily apparent. The boundaries of the trend band reflect the variations resulting from differences in the materials, the irradiation temperature and nuclear environmental conditions. In spite of these variables, a relatively consistent trend has been developed showing the increase in NDT for these two steels.

In addition to the several test reactor irradiation facilities used, data from Charpy V-notch specimens from the pressure vessel of the SL-1 reactor were satisfactorily compared with those from the accelerated irradiation experiments on similar A212-B specimens. The SL-1 specimens received a neutron dose of 1.6×10^{18} n/cm^2 (>1 Mev) with a resultant NDT increase close to that which would be predicted from the trend of Fig. 1 for irradiation at 420°F.

The general agreement established for the NDT data in terms of the 1 - Mev criterion (Figs. 1-6), as well as the agreement for exposure in the highly thermalized neutron environment of the BGR (Fig. 7) and the direct comparison with the operational condition of the SL-1

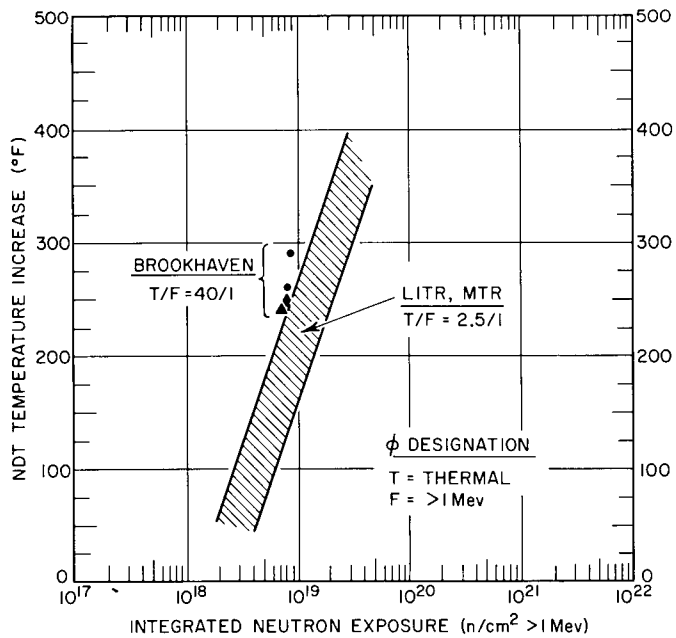

Fig. 8 – Comparison of embrittlement developed by high and low ratios of thermal to fast neutron energy spectra (>1 Mev). The $40/1\phi$ ratio experiment also represents a dose rate of $1/10$ that of the $2.5/1\phi$ ratio experiments included in the band which represents data of Fig. 1.

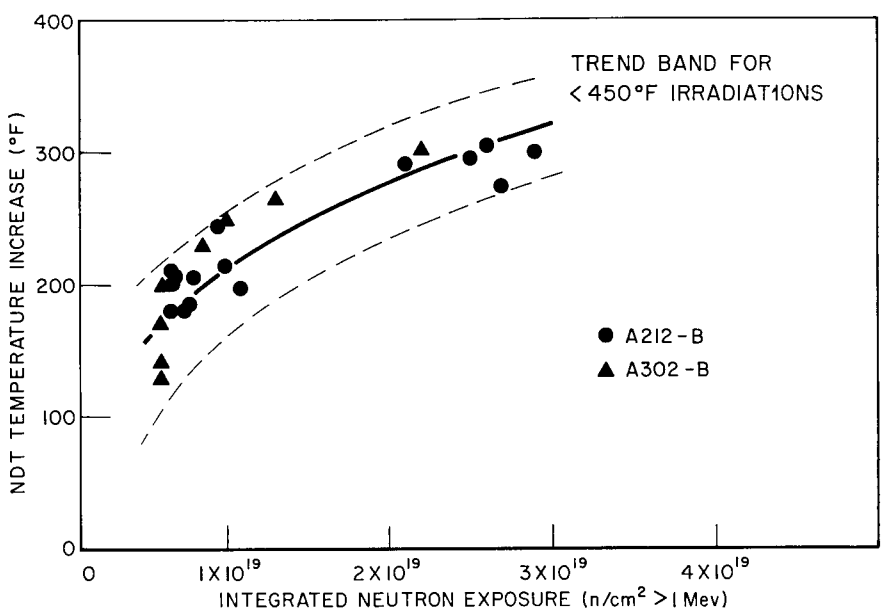

Fig. 9 – Increase in the NDT temperatures of A212-B and A302-B steels resulting from neutron irradiation at temperatures below 450°F. Linear plot shows effect of different neutron exposures.

reactor, demonstrate the practical validity of the 1-Mev reference point. This conclusion is reinforced by the agreement between investigators using this criterion for reporting neutron exposure. Agreement is very good within the United States (7,8), and extends also to European investigations of Harries in the United Kingdom (9), and Weisz in France (10).

The question of rate effect has been studied by Harries, Barton, and Wright in the United Kingdom (11): no effect of neutron exposure rate was observed for the same total neutron dosage accumulated at rates varying from 1 to 100 proportionally and at temperatures between 212° and 660° F. This range of neutron dose rate conditions encompasses the extremes of irradiation rates of NRL experiments and rates anticipated for operating reactors. Thus the data of Harries and co-workers were found to substantiate and extend the conclusions drawn from a comparison of the BGR and MTR-LITR data (Fig. 8). These results will be supplemented further by the surveillance data now available, or soon to be forthcoming, from several commercial nuclear power plants and two Army nuclear stations.

Data from the specimens exposed for long times in nuclear plants will also provide answers to the question of possible neutron energy spectrum effects. However, from data now available, no indications of important neutron spectrum or rate effects are observed. Thus, it appears that the 1-Mev criterion for reporting neutron dosage is valid, at least for the major U. S. nuclear power systems. Furthermore, although the evidence is quite limited, there is no indication that accelerated irradiation data could not be applied to the operating reactor without corrections for the slower rate of neutron exposure in the operating power plant.

Irradiation Temperature – Most of the current U. S. nuclear power plants operate with the pressure vessel temperature in the range from 400° to 600° F. Even the most advanced water systems and the first generation of gas-cooled reactors in the U. S. are not expected to operate with the pressure vessel temperature appreciably higher than 600° F. Nevertheless, the trend for reactor operating temperatures is upward, hence the need for emphasizing irradiation effects at higher temperatures in order to anticipate conditions in reactors yet undesigned and to select the best materials for the expected environment.

Past practice in irradiation experimentation generally involved initial exposure in sealed capsules which usually operated at low temperatures (below 300° F). Such experiments are not complex and permit the rapid accumulation of data for the determination of irradiation effects without significant thermal effects. The range from 400° to 550° F is of special current interest because the early Army and commercial plants which operate in this range have accumulated the largest neutron exposures. While data in this range are growing, the extent of knowledge of neutron embrittlement of steels in and above this temperature range is limited at the present time.

Data in Figs. 1-6 include several data points for irradiation between 400° and 450° F which in general show no appreciable effect of irradiation temperature when compared to irradiation at <300° F. For example, two steels, A212-B and A302-B, were irradiated simultaneously to respective neutron dosages of 6.6×10^{18} and 5.0×10^{18} n/cm^2 (>1 Mev) at four different temperatures: 260°, 400°, 450°, and 550° F. Results of this irradiation are presented in Table 7. From these data, it is seen that no appreciable correction of neutron-induced "damage" is effected during irradiation at temperatures of 450° F or less. The amount of shift in both the A212-B and A302-B materials irradiated at 400° and 450° F is only slightly less than that observed for the 260° F exposure. Irradiation of these materials at 550° F, however, produced a significant effect. The NDT shifts at this temperature are approximately 100 degrees less than those produced at the three lower temperatures.

The above data indicate that the process of annealing of neutron-induced effects during irradiation does not occur in a continual fashion as the irradiation temperature is progressively increased above 200° F. Instead it appears that, somewhere in the range from 450° to 550° F, a thermal annealing mechanism becomes effective.

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Table 7
NDT Increase for Steels Irradiated
at Various Temperatures

Steel	Irradiation Temp. (°F)	Neutron Dosage (n/cm^2: >1 Mev)	ΔNDT (°F)
A-212-B (4″ plate)	260	6.6×10^{18}	210
	400	6.6×10^{18}	180
	450	6.6×10^{18}	200
	550	6.6×10^{18}	100
A302-B (6″ plate)	260	5.0×10^{18}	170
	400	5.0×10^{18}	130
	450	5.0×10^{18}	140
	550	5.0×10^{18}	65

The smaller embrittlement at elevated temperatures is demonstrated by Fig. 10 for irradiation temperatures between 500° and 750° F. These data are presented along suggested trend lines for various temperatures in reference to the trend band for irradiations involving temperatures <450° F. No significant departure from the slope established for low temperature irradiation has been observed. The progressive and appreciable reduction in embrittlement gained through irradiation at these higher temperatures is obvious. Although the benefit of irradiation at successively higher temperatures becomes progressively smaller, the improvement may be of great significance in assessing the effects of high energy neutron irradiation on the structural components of future reactors.

Differences which may be related to the material irradiated at the higher temperatures are not readily apparent, although the separation of data points on the 550° F trend line and the lone point for the A350-LF3 (irradiated at 510° F) suggest compositional or microstructural effects similar to those observed for irradiation at temperatures under 450° F. That is, the A350-LF3 shows greater embrittlement and the HY-80 shows less embrittlement than that observed for the A212-B steel. However, the significance of these materials differences will depend upon additional determinations.

One important anomaly observed with the HY-80 steel at 750° F demonstrates that each pressure vessel steel considered for high temperature nuclear application should be evaluated by advance irradiation at the anticipated operational temperature. The HY-80 steel was irradiated simultaneously with A302-B at three temperatures (540°, 640°, and 740° F) to a neutron dosage of 3.1×10^{19} n/cm^2 (>1 Mev). The results of this irradiation are presented in Table 8. The pattern of successively smaller NDT increases at higher temperatures was broken by the results on the HY-80 at 740° F which showed an unexpected NDT increase of 225° F. A control test to determine the thermal effects for an equivalent 2000-hour period at 740° F produced an NDT temperature increase of only 50° F leaving a "net" irradiation change of 175° F, which is far larger than the shift of 65° F for the A302-B at 740° F. This

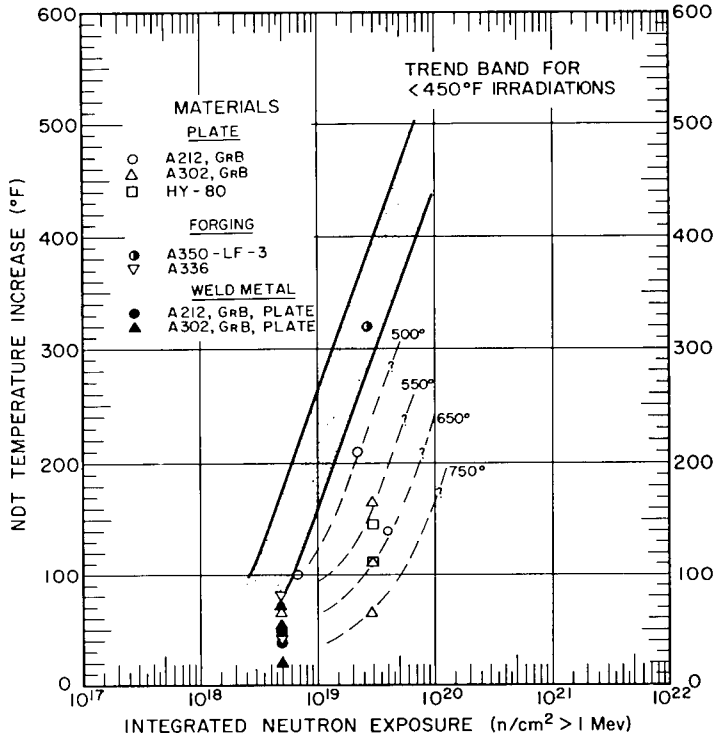

Fig. 10 – Increase in the NDT temperatures of steels resulting from irradiation at temperatures above 450°F. Data points at 5×10^{18} n/cm^2 represent early irradiations in the BGR at 500°-600°F.

Table 8
NDT Increase for Two Steels Irradiated at Elevated Temperatures to Neutron Dosage of 3.1×10^{19} n/cm^2 (>1 Mev)

Steel	Irradiation Temp. (°F)	ΔNDT (°F)
A302-B (6″ plate)	540	165
	640	110
	740	65
HY-80 (Ni-Cr-Mo) (3″ plate)	540	145
	640	110
	740	225

observation is apparently the result of the simultaneous and combined effects of neutron exposure and the elevated temperature operating through a mechanism which is not understood at this time.

The full pattern of embrittlement effects of irradiation of steels at elevated temperatures is not well defined. However, programs now underway at several laboratories will contribute greatly to the knowledge of the effect of this factor.

HEAT TREATMENT OF IRRADIATED STEELS

The maximum embrittlement observed to date is the ΔNDT value of 495° F for the A350-LF3 steel (Fig. 1) after irradiation at 300° F to a neutron dosage of 4.0×10^{19} n/cm^2 (>1 Mev). The observed changes in the A212-B and A302-B steels are smaller. However, these changes are still sufficient to suggest the need for the study of some means for restoration of notch ductility in these steels after irradiation to neutron dosages in the range above about 3×10^{19} n/cm^2(>1 Mev). This need for restoration of notch ductility is especially important for steels irradiated at temperatures below 450° F.

Postirradiation heat treatment techniques have been applied in an effort to provide a recovery of ductility after irradiation embrittlement. The postirradiation annealing studies, which are still in an exploratory stage, have been directed primarily to an evaluation of pertinent variables including (a) the material, (b) the irradiation temperature, and (c) certain factors in the annealing process, especially the duration and temperature of the heat treatment. In addition, preliminary experiments have been conducted to determine the effects of a cyclic irradiation and the annealing process. These data are of practical engineering interest to any decision to anneal the pressure vessel of an operating nuclear power plant.

Effects of Major Variables

Materials – Postirradiation heat treatment studies have included the several steels described in Table 2 with various neutron exposure and annealing conditions. Although no clear pattern regarding the effect of annealing on different steels has been established, a trend has been observed. In general, those steels which show the greatest embrittlement also show the greatest amount of recovery with a given heat treatment. This statement is based upon the results of a single irradiation-annealing experiment in which five steels (Table 4) were exposed to a neutron dosage of 7×10^{18} n/cm^2 (>1 Mev) at 240° F. These steels were then annealed at 600° F for 18 hours. The results of irradiation embrittlement and subsequent recovery through annealing are shown in Fig. 11. These five steels which differ considerably in composition and microstructure are listed in order by increasing nickel content. The extent of notch ductility recovery is directly related to the magnitude of the initial shift in the NDT temperature with the exception of the two very similar A350 steels. These results are not readily explainable but offer some interesting possibilities in connection with engineering application as well as for fundamental studies of radiation damage mechanisms.

Another example of the materials factor in assessing postirradiation annealing behavior involves a comparison between A212-B and A350-LF1 (Mod.) steels. The data presented in Table 9 show the results of annealing at 600° F for 168 hours after irradiation at 430° F. This irradiation temperature is representative of normal operational levels in the Army reactors, SM-1 and SM-1A, which have pressure vessels of A212-B and A350-LF1 (Mod.), respectively. Furthermore, the ΔNDT values listed in this table are in the range which would evoke attention to the possibilities for annealing if the pressure vessel should sustain an equivalent embrittlement. The differences observed may be quite significant in considerations of cyclic irradiation-annealing treatment where the cumulative residual ΔNDT is the critical factor.

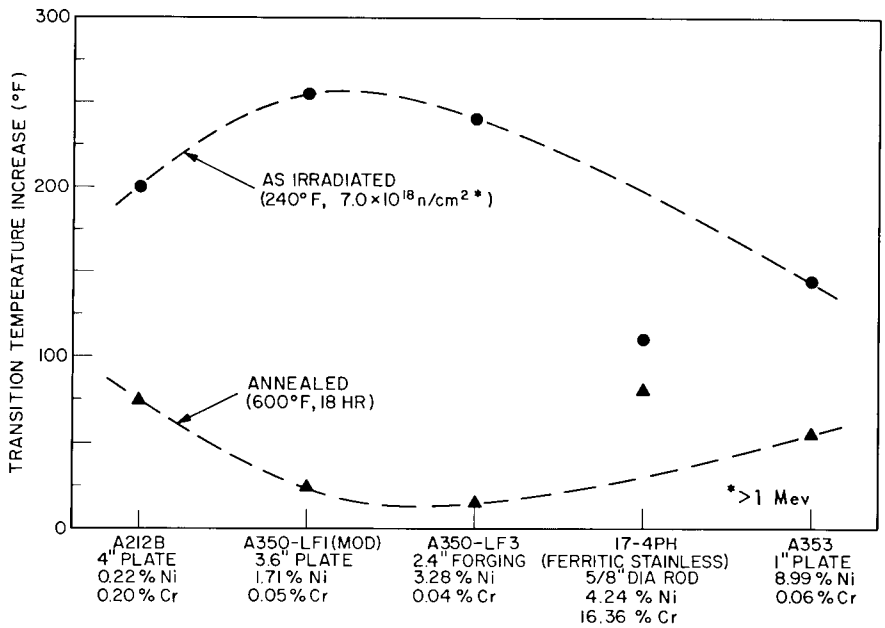

Fig. 11 – Neutron-induced increases in transition temperature for five steels irradiated simultaneously. Lower curve shows restoration of notch ductility through postirradiation annealing.

Table 9
Effects of Postirradiation Annealing on Two Reactor Pressure Vessel Steels (Annealed 168 Hours at 600°F)

Steel	Irradiation Temp. (°F)	Neutron Dosage (n/cm^2 : >1Mev)	ΔNDT (°F)	Recovery	
				ΔNDT (°F)	%
A212-B (4" plate)	430	2.6×10^{19}	305	80	26.2
A350-LF1 (Modified) (3-5/8" plate)	430	3.1×10^{19}	440	345	78.4

Irradiation Temperature – The effect of irradiation temperature on annealing results has been studied also. The results to date indicate two trends which may be considered contradictory and may be related to the materials factor. One trend is evidenced by the results of two experiments with irradiation conditions as follows: one experiment involved exposure of A212-B simultaneously at 200° and 430°F, and a second experiment involved exposure of A212-B simultaneously at 275° and 510°F. These two experiments demonstrate a smaller capacity for annealing recovery after irradiation at the higher temperature. Data from these two experiments are shown in Table 10. In each experiment the same annealing conditions were used for specimens irradiated at both low and high temperatures, and it is apparent that the annealing treatment is much more effective for the low temperature

Table 10
Results of Annealing of A212-B Steel After Simultaneous Irradiation at Low and High Temperatures
(Two Experiments)

Experiment	Irradiation Temp. (°F)	Neutron Dosage (n/cm^2: >1 Mev)	ΔNDT (°F)	Annealing		Recovery	
				Temp. (°F)	Time (Hr)	ΔNDT (°F)	%
A	200	2.5×10^{19}	295	600	168	190	64.4
	430	2.6×10^{19}	305	600	168	80	26.2
B	275	2.1×10^{19}	290	750	36	245	85.6
	510	2.2×10^{19}	210	750	36	135	64.0

irradiation condition. The second trend was observed after annealing A302-B specimens which had been irradiated at three temperatures, 540°, 640°, and 740°F. The results of this experiment, which are presented in Table 11, indicate that an annealing temperature of a given increment above the irradiation temperature results in progressively greater percent recovery as the irradiation temperature is increased. These data would indicate a very marked advantage for the annealing treatment for irradiations at the higher temperatures. More data will be required before the engineering significance of the tendency observed in this experiment can be fully assessed. However, this observation offers encouragement for correcting embrittlement sustained through elevated temperature irradiation of steels in advanced reactor systems.

Table 11
Recovery of Notch Ductility of A302-B Steel
Irradiated at 540°, 640°, and 740°F
(Neutron Exposure - 3.0×10^{19} n/cm^2; energy >1 Mev)

Irradiation Temp. (°F)	ΔNDT (°F)	Annealing Conditions		Recovery	
		Temp. (°F)	Time (Hr)	ΔNDT (°F)	%
540	165	750	24	80	49
640	110	800	24	70	64
740	65	900	24	55	85

The results obtained for the A302-B steel in this experiment were not duplicated for HY-80 irradiated and annealed in a similar pattern. The HY-80 irradiated at 540°F showed a similar degree of recovery on annealing, but after irradiation at 640°F and annealing at 800°F no recovery was observed. This anomalous behavior concurs with the peculiar embrittlement enhancement observed for this steel after irradiation at 740°F. This unusual irradiation behavior indicates the necessity for studying the effects of radiation on each potential reactor pressure vessel steel with an appropriate range of thermal conditions.

Time and Temperature of Annealing – A key factor in any decision to anneal a reactor pressure vessel is the question of annealing conditions to provide an optimum in recovery of properties. The decision to anneal a pressure vessel would be guided by the results of studies of time and temperature effects, but the upper limits of these factors should be established by practical engineering considerations for the particular plant.

A tendency toward re-embrittlement has been observed with prolonged heat treatment (up to 168 hours) of A212-B and A302-B steels irradiated at temperatures below 300°F. That is, after some restoration of ductility with short-term annealing, embrittlement returns as annealing is prolonged. This problem was reviewed in an earlier report (3). Additional experiments have been conducted in an effort to observe a similar "reembrittlement" after irradiation at normal reactor operating temperatures. Fortunately, this phenomenon is not observed with any of the steels studied if the irradiation temperature is 430°F or greater. This means that, for annealing a reactor pressure vessel, consideration of the length of the annealing treatment need not be restricted by concern for a return toward the initial as-irradiated brittleness condition. On the contrary, recent data indicate that recovery proceeds with prolonged treatment at a particular temperature. For example, the percentage recovery of notch ductility on A212-B steel specimens irradiated at 510°F to 2.2×10^{19} n/cm^2 (>1 Mev) showed a marked increase with an extended irradiation period. Recovery amounted to 12 percent with a 600°F treatment for 36 hours but increased to 55-percent recovery after 96 hours at the same temperature. A similar observation was made on the A350-LF1 (Mod.) steel. Table 12 shows the results of various time-temperature combinations for annealing this steel after irradiation at 430°F. Neutron exposure was 3.1×10^{19} n/cm^2 (>1 Mev). The distinct advantage of higher temperatures and longer periods of annealing treatment are vividly demonstrated by the increasing recovery percentage values and the decreasing residual ΔNDT values. A similar pattern has been observed with the A302-B steel as well. Thus, it would appear that a reactor vessel should be annealed at as high a temperature as possible for as long a period as possible to insure maximum correction of neutron embrittlement. This is only part of the answer, however, since the effect of irradiation after one or more irradiation-annealing cycles is all important to the case of an operating nuclear plant. The effect of cyclic irradiation-annealing treatment is being studied to provide answers to this practical engineering problem.

Table 12
Recovery of Notch Ductility of A350-LF1 (Mod.) Steel After Irradiation at 430°F to 3.1×10^{19} n/cm^2 (>1 Mev)
(As-Irradiated ΔNDT = 440°F)

Postirradiation Annealing Conditions		Recovery		Residual ΔNDT (°F)
Temp. (°F)	Time (Hr)	ΔNDT (°F)	(%)	
500	18	55	13	385
500	168	130	30	310
600	1	165	38	275
600	18	285	65	155
600	168	345	79	95
750	1	400	91	40
750	18	425	97	15

Cyclic Irradiation-Annealing Effects

The significant benefits of heat treatment to a reactor pressure vessel must be assessed in terms of not just one irradiation-annealing cycle but multiple cycles. The comparison of cumulative embrittlement with or without annealing must be the determining factor in decisions regarding reactor pressure vessel annealing. If the rate of embrittlement were shown to be greater after irradiation and annealing than in the initial period, the heat treatment would obviously not be worthwhile and might, in fact, be detrimental. Preliminary experiments aimed at determining the effects of cyclic irradiation-annealing treatments have been conducted.

Initial cyclic studies on A350-LF3 and A212-B steels involving irradiation at 240°F and annealing at 700°F indicate that, following an intermediate anneal, the steels investigated were insensitive to their prior irradiation history. In effect, the steels exhibited virgin material characteristics during a subsequent irradiation-annealing cycle. Whether or not this behavior persists with multiple cycles or with elevated temperature exposures remains to be proven. Regardless of the outcome, the assessment of the value of intermediate annealing treatments will continue to be predicated upon the ability of the periodic heat treatment program to measurably reduce the cumulative embrittlement of the steel relative to that developed without intermediate annealing.

An analysis of the relative effect of cyclic irradiation-annealing treatments on an A350-LF3 forged steel is shown in Fig. 12. Since this steel exhibits excellent annealing response (low residual effect) following irradiation at 240°F, the cyclic treatment is very beneficial. Although a cumulative increase in NDT with successive cycles is noted, the projected NDT increase following a third exposure (shaded triangle) would be about 125 degrees less than the increase without intermediate annealing.

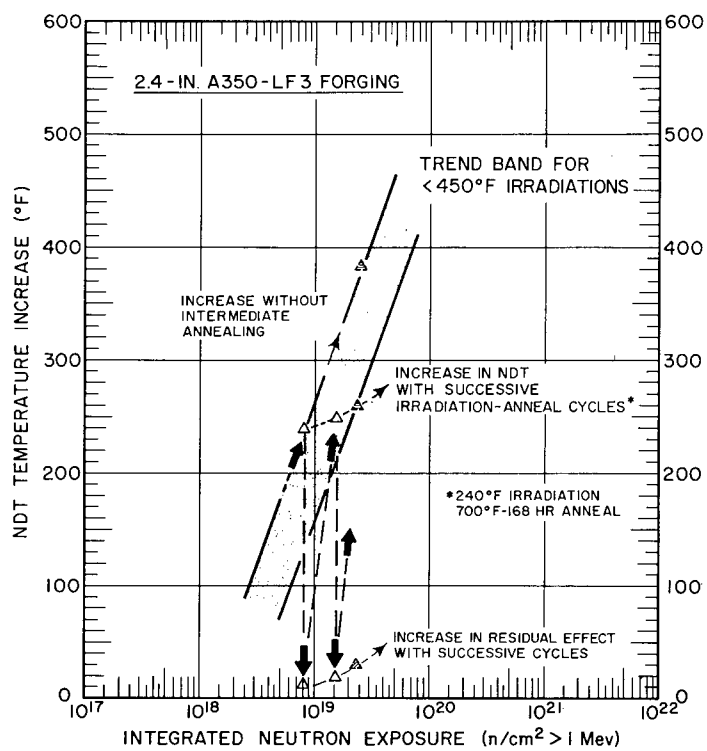

Fig. 12 – NDT temperature behavior exhibited by A350-LF3 forged steel at various stages of cyclic irradiation-700°F annealing treatments

A similar analysis of the notch ductility behavior of an A212-B steel under several irradiation-anneal cycles is presented in Figs. 13 and 14. The poorer recovery characteristics of this steel, depicted by the larger and more rapid increase in residual irradiation effect with successive 600° or 700° F annealing treatments, results in a cumulative NDT increase after the third exposure of only 65 or 70 degrees less than for a comparable irradiation without intermediate annealing treatments. For these exposures (2.1 and 2.3×10^{19} n/cm^2 >1 Mev, Figs. 13 and 14, respectively) on the A212-B steel, intermediate annealing would clearly not be worthwhile since the net advantage in terms of ΔNDT would be masked by the degree of uncertainty as to the actual NDT temperature of the vessel.

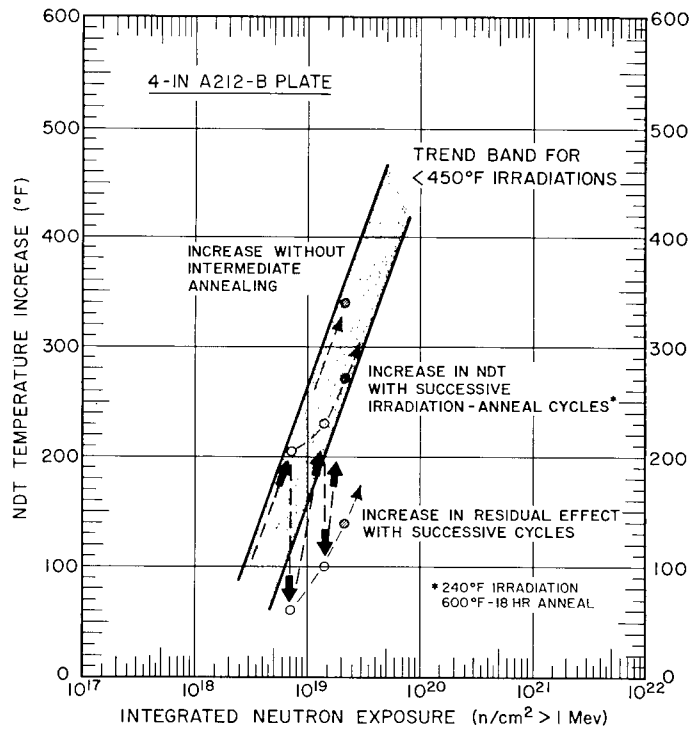

Fig. 13 – NDT temperature behavior exhibited by A212-B steel at various stages of cyclic irradiation-600°F annealing treatments

An interesting comparison may be made between results of the cyclic irradiation-annealing experiments on A212-B (Figs. 13 and 14) and the summary trends for elevated temperature irradiation shown in Fig. 10. If the trend curve for increase in NDT with successive irradiation-anneal (600° F) cycles (Fig. 13) is superimposed on Fig. 10, the trend curve falls between the 550° and 650° F trend lines. Similarly, the trend line for 700° F annealing cycles of Fig. 14 falls between the 650° and 750° F trend lines of Fig. 10. The implication of this observation is that the net ΔNDT after low temperature irradiation-annealing cycles is equivalent to that attained through irradiation at a temperature equivalent to that used for annealing. If this preliminary observation on A212-B steel is proved by further study, the results may be very significant to the planning for minimizing neutron embrittlement in reactor pressure vessels.

Another experiment involved irradiation at a temperature of 650° F and an intermediate in-reactor anneal at 800° F. This preliminary experiment involved the exposure of a

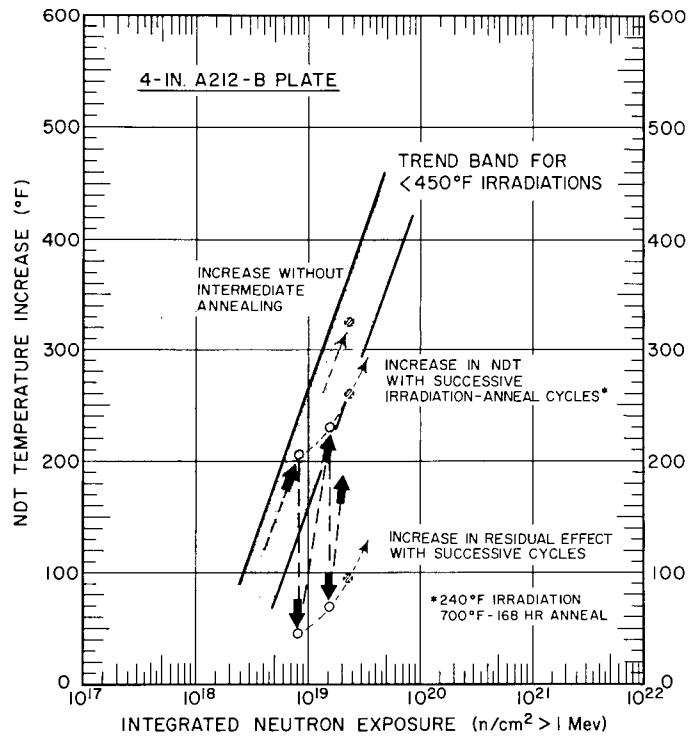

Fig. 14 – NDT temperature behavior exhibited by A212-B steel at various stages of cyclic irradiation-700°F annealing treatments

two-unit assembly in the LITR for five reactor cycles. The intermediate annealing was accomplished during reactor operation by the addition of the heat generated by a small furnace in one zone of the two-unit assembly. The in-reactor thermal history of this assembly is shown in Fig. 15. The lower zone, which contained the reference or control material, operated over the whole irradiation period at temperatures lower than the top (annealed) zone. In spite of imperfect temperature control, an advantage of intermediate annealing is shown (Table 13). Assuming no major effect due to the differences in irradiation temperature, the benefits of intermediate annealing under these conditions is significant, since this recovery was effected after a relatively small total neutron dose. It would appear that the second cycle tendency toward embrittlement at the rate for virgin material as observed with low temperature irradiations may not be effective for the elevated temperature cyclic experiment. Consequently, if the observed net advantage of intermediate annealing applies with subsequent annealing cycles, this technique may be useful for reducing pressure vessel embrittlement. However, before broad conclusions can be reached, much remains to be explored experimentally, particularly the relationship between annealing response and irradiation temperature, and the quantitative effect of prior neutron dosage accumulation on the residual NDT increase after annealing.

APPLICATION OF EXPERIMENTAL OBSERVATIONS TO NUCLEAR POWER REACTOR CONDITIONS

Surveillance of Operating Nuclear Plants

In addition to the question of possible differences in neutron embrittlement of steels as a result of differences in nuclear environmental conditions between test reactors and power

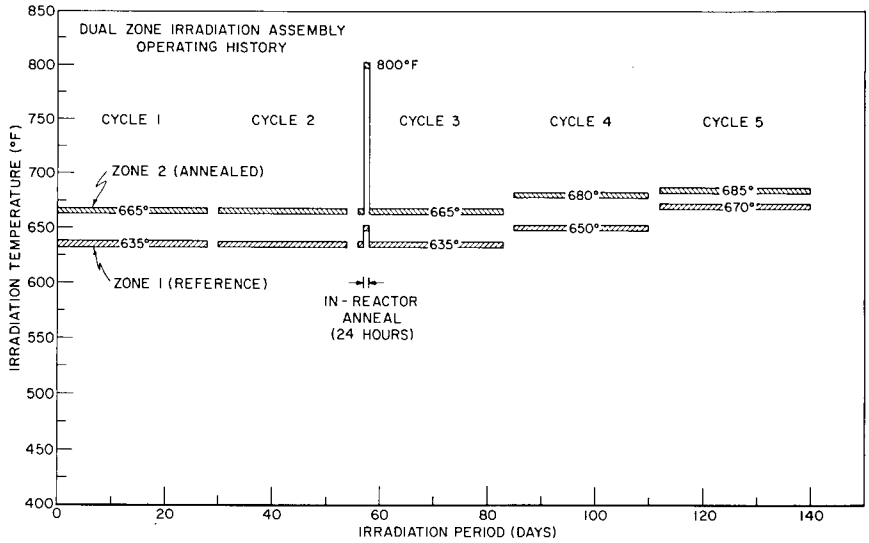

Fig. 15 – Thermal history of in-reactor cyclic irradiation-annealing experiment on A212-B steel

Table 13
Effects of Cyclic Irradiation-Annealing on Notch Ductility of A212-B Steel
(Irradiated at 650°F to Approximately $4 \times 10^{19*}$ n/cm^2: >1 Mev)

Experiment	In-Reactor Anneal	ΔNDT (°F)
Reference Zone	None	140
Annealed Zone	800°F, 24 hours	105

*Annealed after exposure of approximately 1.6×10^{19} n/cm^2 with subsequent additional exposure of about 2.4×10^{19}.

reactors, there is the important question of the effects of the stressed condition of the operating pressure vessel. The nuclear environmental aspects, neutron dose rate and neutron energy spectrum have been discussed. The primary effort to resolve these questions involves programs for placing representative specimens of critical reactor structural components inside operating power reactors. Radiation damage surveillance programs devised for commercial reactors have been described by DiNunno and Holt of the USAEC (5). Programs now underway in two Army reactors have been described by the authors (12,13). Some results of surveillance in the Dresden reactor have been reported by Brandt and Alexander (8). The surveillance program for the Yankee Atomic Plant has been described by Landerman (14).

The preliminary pressure vessel surveillance data from the Dresden plant (A302-B vessel) agree well with NRL experimental data on the same steel, thus indicating no major effects of neutron dose rate or spectrum. No other power reactor surveillance data are available at this time; however, data from specimens to be removed from the Army reactors, SM-1 and SM-1A, will be available during 1963.

Another facet of the power reactor surveillance program involves the placement of neutron dosimeters along the inside of the pressure vessel wall. A particularly promising dosimetry technique for this application is the activation reaction $Fe^{54}(n,p)Mn^{54}$ since the product has a long half-life (314 days) and the effective neutron threshold for this reaction is satisfactory. The authors have suggested that, with broader experience of correlating embrittlement with neutron exposure determined by this reaction, it may be possible to assess radiation damage through the Mn^{54} dosimetry technique (15). This technique may permit surveillance without the placement of specimens in an operating plant and may also be applied to steel reactor components that are taken out of service. In fact, the Mn^{54} constituent in the SL-1 reactor pressure vessel was used to determine neutron exposures in the belt line (peak flux) portion and thus permitted correlation with data from accelerated irradiation experiments.

At the present stage of knowledge, however, it is preferred that both specimens and neutron dosimeters be applied in power reactor surveillance programs so that a direct correlation with other experimentally observed data can be obtained. A program of this type is being conducted by NRL in the Army SM-1A reactor at Ft. Greely, Alaska. The first phase of the effort, neutron dosimetry along the inside pressure vessel wall, has been completed.

Figure 16 presents schematically the details of the major critical region covered by the neutron flux survey and the instantaneous neutron flux values for specific points along the inside of the pressure vessel wall. Average reactor power output during the activation of dosimeters was 7.1 megawatts compared with a design full power output of 20.0 megawatts. Within certain limits, the data shown may be used to estimate future embrittlement of the SM-1A pressure vessel. It is apparent that such estimates cannot be optimistic since the peak integrated pressure vessel neutron dosage is $7.8 \times 10^{18} n/cm^2$ (>1 Mev), for one year of full time, full power operation. In Fig. 2, an NDT increase of more than 200° F may be estimated for the irradiation of the SM-1A steel to this neutron dosage at 430° F. However, the full significance of an exposure of this magnitude will be tested when specimens and associated dosimeters from the SM-1A reactor are evaluated. Similar data from the surveillance of other power reactors should further clarify the relationship between radiation damage in test reactors and that in power reactors.

Materials from Power Reactors No Longer in Service

The dismantling and sectioning of the pressure vessel of the SL-1 reactor provided an excellent opportunity for comparison of the SL-1 embrittlement with data on the embrittlement of other reactor steels which have been obtained through accelerated irradiation experiments in test reactors.

The normal operating temperature and pressure of the SL-1 reactor were 420° F and 300 psig. The nominal hoop stress on the 3/4-inch-thick pressure vessel was approximately 11,000 psi during normal operation. During the excursion, however, the hoop stress rose to an estimated 18,000 psi in the central core region. Pressure vessel stress levels were apparently much higher above and below the core region as a result of channeling effects of the core structure, control rod housings, and the thermal shield.

Tension specimens and Charpy V-notch specimens were cut from various sections of the SL-1 pressure vessel for evaluation by several laboratories. Two groups of specimens, one group from the steam drum region and one from the core region, were received by NRL for evaluation. Specimens from the low flux (steam drum) position served as control specimens since the thermal and stress conditions were essentially the same as for the high flux (core region) position. The core region neutron dosage as determined by an analysis for the Mn^{54} produced by the reaction $Fe^{54}(n,p)Mn^{54}$ was $1.6 \times 10^{18} n/cm^2$ (>1 Mev) (16).

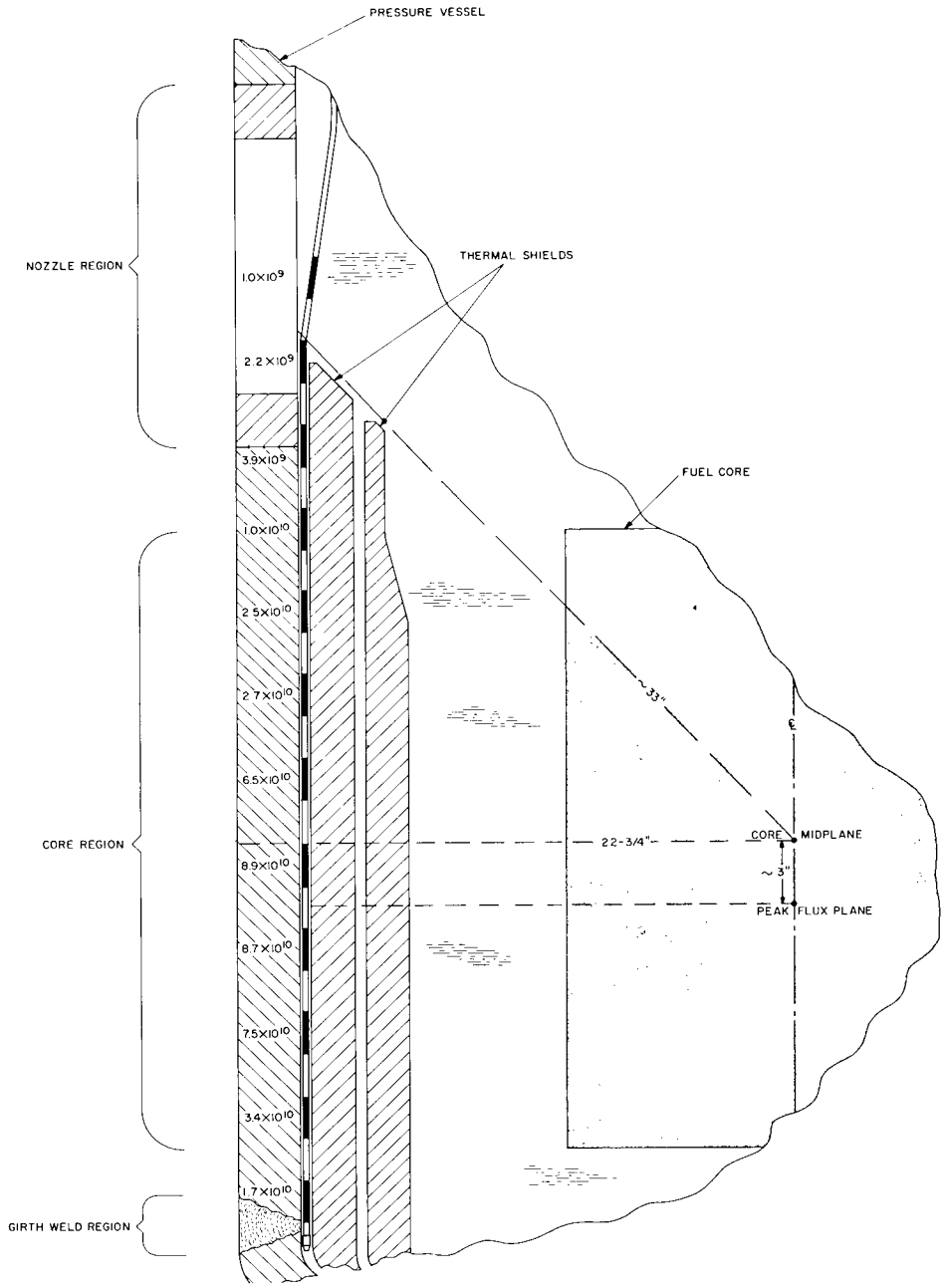

Fig. 16 – Schematic view of critical pressure vessel regions and instantaneous neutron flux values for various positions along the pressure vessel. (Flux based on activation of 304 stainless steel tubing during reactor operation at average power output of 7.1 megawatts.)

The NDT temperature increase for the SL-1 pressure vessel along with other points for accelerated irradiation of specimens of a 4-inch A212-B steel are plotted on the trend band for <450° F irradiations in Fig. 17. The observed increase in NDT for the SL-1 pressure vessel, approximately 70° F, is slightly greater than might be predicted by extrapolation of data from accelerated irradiations of the 4-inch A212-B steel. The difference at this low neutron dosage is so slight, however, that it is not possible to conclude that the effect of applied stress during irradiation is significant. By the same token, it cannot be said that the rate of irradiation or the neutron energy spectrum involved influenced significantly the embrittlement of the SL-1 pressure vessel.

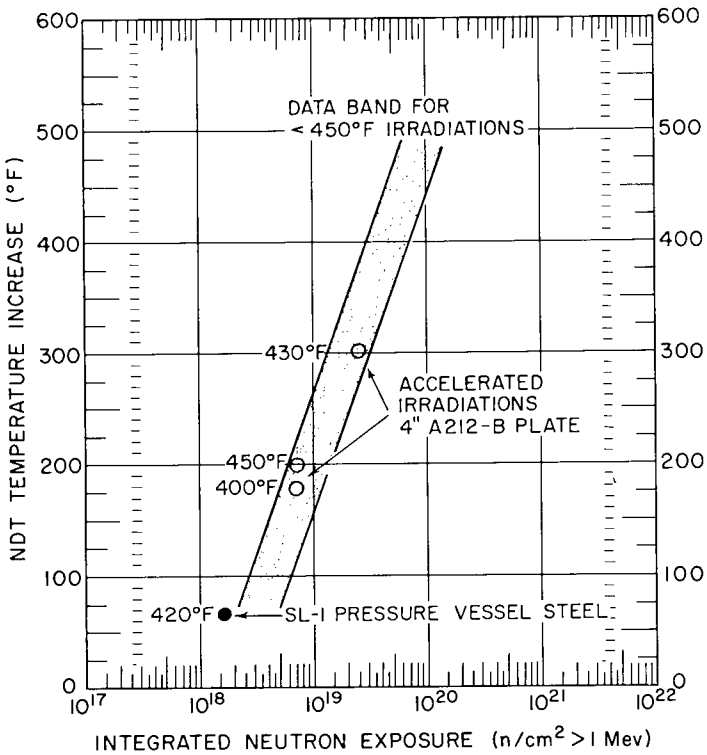

Fig. 17 – Increase in the NDT temperature of the SL-1 reactor pressure vessel compared with increases observed for other A212-B steels irradiated at similar temperatures. Data points are shown with reference to the trend from Fig. 1.

Another opportunity for the evaluation of materials from an operating reactor is presented by the recent terminal shutdown of the Organic Moderated Reactor Experiment. The USAEC has requested NRL to take dosimetry samples from various portions of the pressure vessel and other structural components and to evaluate the neutron exposure sustained by these members. These data will permit an evaluation of anticipated changes in the properties of the steel in view of current experimental data. A decision as to the desirability of more comprehensive studies involving the removal and evaluation of the properties of specimens from portions of structural components will depend upon whether or not the magnitude of the neutron dosage determined is sufficient to indicate appreciable changes in the mechanical properties.

SUMMARY AND CONCLUSIONS

The report is intended to be a status report on NRL studies of neutron embrittlement of reactor steels. Accordingly, although there are many points which are not yet clear, certain conclusions can be made firmly and others with reservations. The following list of conclusions is presented to indicate the status of the understanding of neutron embrittlement of reactor pressure vessel steels in terms of the major experimental factors.

1. Fracture analysis of structural steel failures has demonstrated a point of relative safety from brittle fracture at a point 60° F above the nil ductility transition (NDT) temperature.

2. The NDT temperature of reactor pressure vessel steels, before and after irradiation, may be determined by examination of the Charpy V transition curve if an NDT reference point has been established by direct comparison to results of the drop weight test. (The 50-percent shear reference point method is used to determine the NDT point when the ductile portion of the Charpy curve drops below the reference energy level for the unirradiated steel.)

3. To date the difference in the degree of neutron embrittlement observed between the several steels studied are not considered significant in terms of nuclear engineering. Nevertheless, differences between the A350 steels, which show the greatest embrittlement, and the HY-80 and A353 steels, which show the smallest embrittlement, are sufficient to encourage the possibility of finding steels with minimum radiation sensitivity. Some differences in irradiation response have been observed for both low and elevated irradiation temperatures.

4. The validity of the >1 Mev energy criterion for establishing the effective neutron dosage has been verified by the data trends which are consistent in spite of the variety of reactors used for the irradiation experiments.

5. There is no indication that the data obtained in accelerated test reactor experiments cannot be applied directly to the condition of the operating nuclear reactor for reasons of differences in the neutron dose rate or energy spectrum. The results of several experiments as well as the evaluation of specimens from the SL-1 reactor pressure vessel give rise to this conclusion. Power reactor surveillance experiments will contribute valuable data for resolving this question more fully within the next few years.

6. For all practical purposes, the degree of embrittlement of steels irradiated in the range of 200° to 450° F is independent of the temperature.

7. With successively higher irradiation temperatures in the range from 450°-750° F, the degree of embrittlement is progressively smaller. One exception to this observation is the HY-80 steel irradiated at 740° F which showed enhanced embrittlement relative to irradiation at 540° and 640° F. This effect is apparently related to some phenomenon involving combined thermal and neutron irradiation effects on this particular steel. This observation indicates the necessity for determining the effects of neutron irradiation on each steel which is considered for nuclear application at elevated temperatures.

8. The extent of recovery of notch ductility of steels through postirradiation heat treatment is shown to depend upon the following factors:

a. The Material. Those steels which show the greatest embrittlement tend to show the greatest recovery for specific experimental conditions.

b. The Irradiation Temperature. If the annealing temperature is a specific increment, for example 150° F, above the irradiation temperature, the percentage recovery is progressively greater for the higher irradiation temperature.

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c. Time and Temperature of Annealing. For steels irradiated in the normal temperature range of reactor operation, 400° to 600°F, the degree of recovery depends upon both the duration and the temperature of annealing. The major recovery determinant is the temperature. The recovery process proceeds at a diminishing rate with time.

d. The Neutron Exposure. There is some indication that frequent annealing is more beneficial than infrequent treatment. However, data indicating the effects of neutron dosage accumulation are sparse. Experiments are now underway to help clarify this point.

9. Cyclic irradiation-annealing experiments indicate that, for low irradiation temperatures, the residual ΔNDT after annealing depends upon the steel being studied. The benefits of this treatment are significant for the A350-LF3 steel but are much less beneficial for the A212-B steel. A preliminary cyclic irradiation-annealing experiment on A212-B steel irradiated at 650°F and annealed at 800°F shows a significant reduction in the ΔNDT, in comparison to control specimens which were irradiated under similar conditions but did not receive the intermediate anneal. The results of multiple cycles are required for development of firm conclusions as to the value of pressure vessel annealing for the restoration of notch ductility.

10. Data from the SL-1 reactor pressure vessel, which was irradiated in a stressed condition, show no effect attributable to the stress. However, the determination of the effects of stress on radiation damage accumulation will require much additional effort for clarification.

ACKNOWLEDGMENTS

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The personal contributions of each member of the staff of the Radiation Operations Section are also acknowledged with gratitude. Without a concentrated team effort by this group the necessary accumulation of the neutron irradiation effects data could not have been accomplished.

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Appendix A

EXPERIMENTAL IRRADIATION TECHNIQUES

Encapsulation for Neutron Irradiation

The guiding principles for the preparation of experimental assemblies are: (a) simplicity (for fabrication, for insertion, for control, and for postirradiation disassembly), (b) provision for a large number of specimens under specific conditions, and (c) provisions for knowledge of the thermal and neutron environments during irradiation.

Irradiation experiments have been carried out in five test reactors, the Argonne CP-5 Reactor (CP-5), the Brookhaven Graphite Reactor (BGR), the National Reactor Test Station – Materials Testing Reactor (MTR), the Oak Ridge Low Intensity Test Reactor (LITR), and the Oak Ridge Research Reactor (ORR). In addition, capsule irradiation units have been exposed in the two Army reactors, the SM-1 (stationary, medium power, model 1) and the SM-1A (stationary, medium, model 1A).

The very wide differences in the physical and nuclear characteristics of these seven reactors has dictated a variety of experimental assemblies. Nevertheless, with the exception of the CP-5 and BGR experiments, which were quite limited in scope and involved air cooling, two general types of encapsulation were utilized. These two involved stainless steel sheath capsules (no control instrumentation) and similar capsules with instrumentation. Of course, the physical size and shape of the experimental assemblies varied somewhat from reactor to reactor and in accordance with specific experimental objectives.

Capsule type experiments have been utilized exclusively in the MTR, the ORR, and the two Army reactors. This type of unit has been used to establish data trends at low irradiation temperatures in the MTR and ORR and for irradiation at operating (coolant water) temperature in the Army reactors. Figure A1 shows a typical capsule for irradiation in the MTR. Capsules for the ORR are similar in design and construction. An irradiation array with four attached units for exposure in the SM-1A reactor is shown in Fig. A2 along with two individual capsules. The capsules used in the Army reactors have been described in considerable detail in an earlier publication (1).*

All capsule irradiation units are basically the same. Steel specimens are positioned in an aluminum or stainless steel skeletal framework so that as much contact as possible is maintained between the thin covering stainless sheath and the specimens. The arrangement enhances heat transfer and thus minimizes the temperature during irradiation. The stainless steel sheath is welded around the assembly. The whole unit is then compressed with helium at 1500 to 2000 psi to force the sheath into the configuration of the specimens and to indicate any leaks that are present. The unit is then heated to a temperature approximating the anticipated exposure temperature with a continuous flow of helium through the assembly. While still hot the unit is sealed by welding the helium tube. This compact assembly contains a relatively large number of steel specimens in a limited area, thus providing a specific, common, nuclear exposure condition.

*L. E. Steele and J. R. Hawthorne, "Surveillance of Critical Reactor Components to Assess Radiation Damage," NRL Report 5830, Sept. 1962.

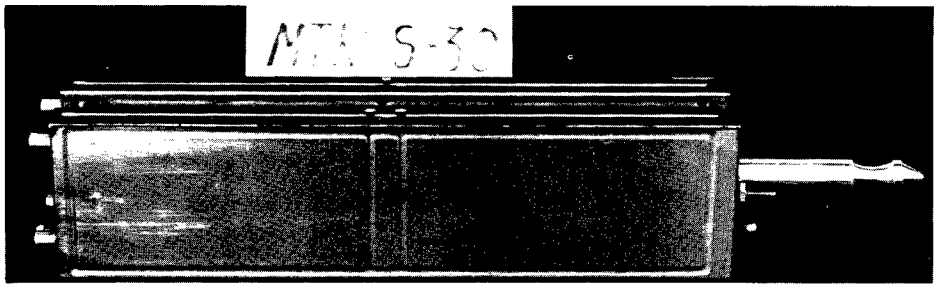

Fig. A1 - Typical capsule for irradiation in the Materials Testing Reactor

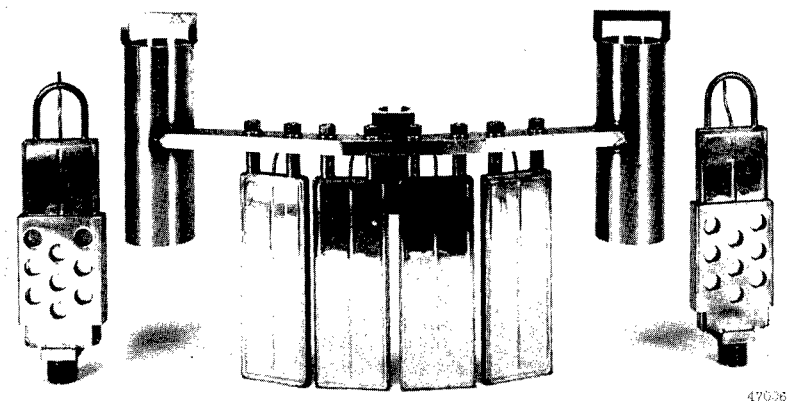

Fig. A2 - SM-1A reactor pressure vessel surveillance specimen assemblies. The curved assembly is mounted above and to the side of the core. The individual units fit in peripheral positions near the top corners of the core.

The instrumented assemblies have involved a much wider range of design considerations. However, these may be considered to be of two general types: (a) assemblies which depend upon gamma heating and variations in heat transfer paths, and (b) assemblies which incorporate furnaces for the control of temperature in the reactor. The first type of assembly has been described previously.* A typical assembly of this class is shown in Fig. A3. An assembly incorporating heaters is shown before encapsulation in Fig. A4.

Simplicity in the control system is the primary advantage of the assembly which depends upon heat transfer variations for temperature control. Control is accomplished through an on-off internal pressurization scheme which forces the sheath away from the specimens cyclically, depending upon thermal conditions. This system is limited to application at relatively low temperatures and has the further disadvantage of being relatively inflexible

*L. E. Steele and J. R. Hawthorne, "Effect of Irradiation Temperature on Neutron-Induced Changes in Notch Ductility of Pressure-Vessel Steels," NRL Report 5629, June 1961.

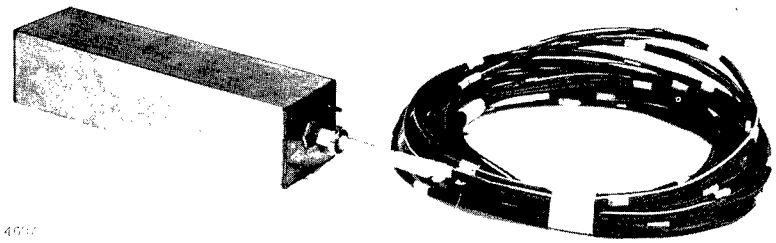

Fig. A3 – Typical assembly for controlled temperature irradiations. Temperature control depends upon internal pressurization to change the heat transfer characteristics of the assembly.

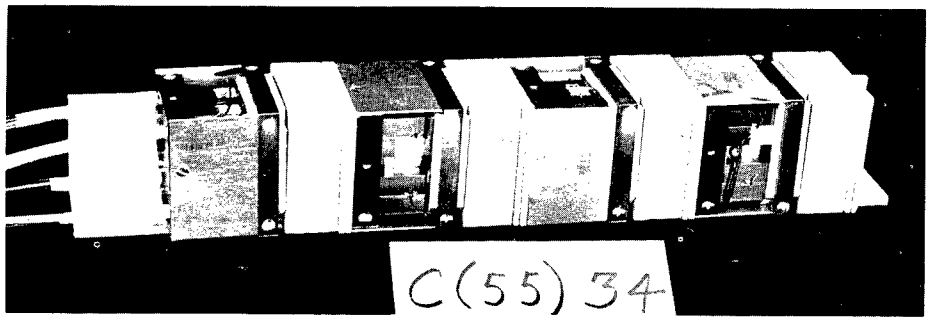

Fig. A4 – Irradiation assembly containing four individually controlled heaters (lighter sections) before encapsulation. This assembly has been utilized in core positions of the LITR.

with regard to the possibilities for changing the desired control temperature during the irradiation period. In order to overcome these disadvantages, in-reactor furnaces have been designed and utilized in several LITR irradiation experiments.* The addition of furnaces has complicated the irradiation control instrumentation but adds considerably to the flexibility for experimental temperature control and greatly extends the range for routine exposure temperature operation.

Temperature sensing and control has been based upon the use of a large number of chromel-alumel thermocouples welded to selected specimens throughout the assembly. Thermocouple reliablility has been excellent. In capsule or non-lead experiments, the maximum temperature attained during irradiation is determined through the use of low melting point metals, alloys, and eutectics.

Most of the irradiation assemblies are exposed in the core lattice of the LITR and the MTR and, accordingly, are limited in cross section by the space inside a dummy fuel piece (approximately 2-1/2 × 2-1/2 inches square with allowance for a sufficient coolant water annulus). The length is effectively limited by the relatively uniform neutron flux region

*J. R. Hawthorne, J. R. Reed, and R. F. Bryner, "In-Reactor Furnaces and Associated Control Instrumentation," Report of NRL Progress, pp. 35-37, Mar. 1963.

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about the center of the fuel element length. This useful length is about 12 inches. These limitations establish the general physical size and shape of over 90 percent of the irradiation units used. Other assembly designs have been required for use in the various facilities of the LITR, BGR, and ORR.

Neutron Dosimetry

One of the most important parameters in the determination of neutron irradiation effects in steel is the measurement of the nuclear environment, especially the flux and dosage of high energy neutrons. This factor has been one of the most difficult in the assessment of radiation damage and, while relative values from experiment to experiment are believed to be excellent, the knowledge of neutron exposure in an absolute sense presents difficulties related to uncertainties regarding the nuclear characteristics of the activation or threshold type detectors normally used for measuring neutron conditions. Discussions of the problem of high energy neutron dosimetry in the study of radiation effects in materials have been presented in other reports.*†‡

Current dosimetry practice, while not ideal, is considered to be adequate for current irradiation studies. The techniques utilized involve the simultaneous irradiation of activation dosimeters with steel specimens in each irradiation experiment. Dosimeters are placed in the notches of Charpy V-notch specimens during irradiation. The high energy neutron dosimeters are high purity iron, nickel, and titanium wires (0.020-inch diam). The reactions involved are $Fe^{54}(n,p)Mn^{54}$, $Ni^{58}(n,p)Co^{58}$ and $Ti^{46}(n,p)Sc^{46}$. The dosimeter for thermal neutrons is cobalt (as a minor constituent in aluminum wire) and for epi-thermal neutrons, cobalt shielded with 0.040-inch-thick cadmium tube.

After irradiation the activated wires were analyzed by the Phillips Petroleum Company, Physics Group, Materials Testing Reactor, Idaho, for the products indicated above, and by application of an average cross-section value for the entire neutron spectrum and the assumption of a slightly moderated fission spectrum, a value in terms of neutron energies above 1 Mev was calculated. The validity of these dosimetry techniques has been demonstrated by a good correlation of dosimetry and notch ductility data from a large number of experiments.

Thermal and epi-thermal flux values are useful in comparing the results of irradiation under different nuclear spectrum conditions and are necessary for making corrections for the burn-out of Co^{58} in a high thermal neutron flux.

Postirradiation Evaluation

All phases of the radiation effects study were conducted by the same group at NRL. The design of irradiation assemblies, which is based largely upon experimental objectives and the limitations of the test reactor, is also influenced greatly by limitations of hot laboratory operations. This factor re-emphasizes the necessity for simplicity in experimental design.

*W. S. Pellini, L. E. Steele, and J. R. Hawthorne, "Analysis of Engineering and Basic Research Aspects of Neutron Embrittlement of Steels," NRL Report 5780, Apr. 1962.
†L. E. Steele, "Practical Neutron Dosimetry for Steel Irradiation Studies," pp. 328-342 in "Steels for Reactor Pressure Circuits," London, The Iron and Steel Institute (1961).
‡C. H. Hogg and L. D. Weber, "Fast-Neutron Dosimetry at the MTR-ETR Site," ASTM Symposium, Radiation Effects on Metals and Neutron Dosimetry, Los Angeles, Oct. 2-3, 1962.

Postirradiation evaluation of irradiated steel specimens for notch ductility has involved two general types of operations: machine shop operations, and impact testing operations. In the machine shop cell, specialized remote tools are used to disassemble, separate, and identify radioactive specimens and neutron dosimeters as well as low melting point thermal detectors in the case of capsule irradiations. The other cell contains the remotely operated drop weight test machine and Charpy impact machine and equipment for thermally conditioning irradiated specimens. In addition, this cell contains furnaces for postirradiation heat treatment studies. The remote impact testing equipment has been described separately.*

The irradiated impact specimens are thermally conditioned in liquid baths or in an air stream with temperature measurement by thermocouple on a control specimen or in physical contact with the specimen to be tested. A minimum of six drop weight test specimens are used to determine an NDT temperature value, and between twelve and sixteen Charpy V-notch specimens are used to establish the Charpy curve. The fracture surface of each broken Charpy specimen is observed to assess the percentage of ductile (shear) fracture versus brittle (cleavage) fracture. Hardness values are also obtained for the Charpy specimens. Annealed Charpy specimens are conditioned and tested on the same equipment as the irradiated specimens.

*L. E. Steele and J. R. Hawthorne "A Remotely Controlled Drop-Weight Test Machine for Brittle-Fracture Studies," NRL Report 5278, Feb. 1959; J. R. Hawthorne and L. E. Steele, "A Remotely Operated Charpy Test Machine for Radioactive Specimens," NRL Report 5305, July 1959.

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