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New Information on Neutron Embrittlement and Embrittlement Relief of Reactor Pressure Vessel Steels

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CONTENTS

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ABSTRACT

Significant embrittlement of several reactor pressure vessel steels as a result of exposure to neutrons has been demonstrated by studies at the U.S. Naval Research Laboratory. Neutron embrittlement of the carbon and low-alloy steels investigated has been defined in terms of increases in the nil ductility transition (NDT) temperature. Increases in the NDT of pressure vessel steels as great as 545°F have been observed. The extent of embrittlement has been shown to depend upon the neutron exposure ($n/cm^2 > 1$ Mev), the type of steel, and the irradiation temperature.

Embrittlement relief through annealing heat treatment at temperatures above the pressure vessel operating temperature has been demonstrated. Significant embrittlement relief was observed even with multiple irradiation-annealing cycles, the extent of relief being primarily dependent upon the irradiation temperature and the subsequent annealing temperature.

Experimental data are reviewed with consideration for application to operating nuclear pressure vessel conditions. Preliminary results of power reactor surveillance and of the evaluation of one pressure vessel after nuclear service are related to experimental results. The value of extending the evaluation of surveillance specimens and of pressure vessels after removal from nuclear service are reviewed with reference to current uncertainties as to possible nuclear environmental and stress effects.

Possibilities for using the favorable aspects of certain variable experimental factors, such as differences in embrittlement sensitivity between steels, are suggested for minimizing steel embrittlement in future reactors.

PROBLEM STATUS

This is a summary report of progress to date; work on several phases of the problem is continuing.

AUTHORIZATION

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NEW INFORMATION ON NEUTRON EMBRITTLEMENT
AND EMBRITTLEMENT RELIEF
OF REACTOR PRESSURE VESSEL STEELS*

INTRODUCTION

Much has been written in recent years about the neutron embrittlement of reactor pressure vessel steels. These reports have demonstrated trends in embrittlement with neutron exposure which involve large increases in the nil ductility transition (NDT) temperature with relatively moderate neutron dosages. When the demonstrated NDT increases are related to applied nuclear engineering within the framework of the comprehensive fracture analysis concept evolved by Pellini and co-workers (1), the implications for potential brittle fracture hazards are quite severe. Accordingly, clarification of the effects of several variable factors including the pressure vessel steel type, the nuclear environment, the thermal environment, and the stress state is essential in order to avoid generalizations which may be unrealistically restrictive in assessing neutron embrittlement hazards for individual reactor systems. This report is devoted to clarifications made possible by new irradiation data for several steels after exposure at various temperatures, and by information regarding the possibilities for embrittlement relief through postirradiation or intermediate heat treatment. In addition, certain anomalous results are reviewed for suggestions regarding future developments in the use of carbon and low-alloy steels in nuclear systems as well as certain implications for understanding the mechanisms of radiation embrittlement. In order to present the new information in proper perspective, however, certain background data are also presented. Thus, this report summarizes the current status of the NRL study.

In this study, emphasis has been placed upon the commonly used pressure vessel steels, ASTM A212-B and A302-B, at temperatures from 200°F to ~750°F. However, several other steels including ASTM A350-LF1 and LF3, HY-80, T-1, and others have been irradiated for information on comparative embrittlement. The observed variable response of different steels and the effects of temperature are reviewed to demonstrate variations under specific neutron dosage conditions.

Because the extent of steel embrittlement observed has been quite large, a study has been undertaken to assess the possibilities of embrittlement relief by means of thermal annealing cycles. Recent results of this phase of the NRL study including results of multiple irradiation-annealing cycles simulating in-service annealing are presented. Effects of the several variable factors, again including the nuclear exposure, the material, and exposure temperature, as well as annealing duration and temperature, are reviewed to show the role of each in the process of neutron embrittlement relief.

The applicability of data from experimental studies conducted in test reactors to the nuclear power reactor condition remains an important question. However, some new information is available from surveillance programs and from specific reactor experiments which is applicable to various questions regarding the nuclear environment. New possibilities for studying these questions are offered by the availability of pressure vessels from reactors such as the Army SL-1 and PM-2A reactors and the Organic

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Moderated Reactor Experiment (OMRE), which have been removed from active service. Preliminary plans for extending the correlation of data from these several new sources are reviewed.

FRACTURE CRITERIA AND IMPLICATIONS FOR NUCLEAR ENGINEERING

In order to produce meaningful notch ductility data for assessing the embrittling effects of nuclear radiation, it is necessary that a relatively simple, yet reproducible, test be conducted to serve as a reference for comparison of the nonirradiated and the irradiated steel transition temperatures. The NDT drop-weight test for determining the nil ductility transition (NDT) temperature (ASTM Recommended Practice E-208-63T) (2) meets these requirements but necessitates the use of relatively massive specimens which are difficult to irradiate under controlled conditions. The other alternative (and the current general practice) involves the use of Charpy V-notch specimens which have been previously correlated to an NDT temperature established by drop-weight tests. Thus, a specific reference transition temperature is determined rather than a transition temperature range which might extend over 100°F or more. The validity of this correlation between the two test procedures has been tested for nuclear environmental effects to neutron exposures as great as 3×10^{19} n/cm^2 (>1 Mev) (3). With proper correlation of Charpy V and drop-weight tests, it is then possible to establish a neutron-induced increase in NDT through the testing of irradiated Charpy V-notch specimens. This procedure has been used for the several steels which have been studied in the NRL irradiation effects program, and most of the irradiated NDT values presented here are based upon Charpy specimens.

With the correlation of notch ductility tests established, the broader significance inherent in the NDT determination becomes more directly applicable to the study of neutron embrittlement of steels. That is, the common frame of reference, NDT, permits the extrapolation of recently developed general concepts of fracture analysis (1) and fracture-safe engineering design (4) to nuclear engineering.

The conditions for brittle fracture initiation have been stated by Pellini and Puzak as follows (1):

> "Brittle fractures may be initiated at conventional design levels of nominal elastic stress, provided certain other conditions are satisfied, as follows: (a) a flaw such as a crack or sharp notch is present, (b) the stress is of sufficient intensity to develop a small amount of deformation at the notch tip, and (c) the service temperature is low enough to promote cleavage fracture of the deformed metal crystals at the notch tip."

They further explain that, "if cleavage cracking occurs, a sharp natural crack front is extended into the metal by a high-speed repetition of the crack tip cleavage process, resulting in a 'propagation' of the brittle fracture."

The analysis of brittle failure of steel structures at NRL has thus emphasized knowledge of three key factors: flaw, stress, and temperature conditions. The analysis of a large number of nonnuclear, steel structural failures in terms of these factors has culminated in the development of a comprehensive Fracture Analysis Diagram which relates these factors over a range of conditions.

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 initiation, even with large flaws present. 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.

Since there is no practical way to assure that flaws of significant size (inches) are not present in operating reactor pressure vessels, 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. 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.

Good knowledge of the NDT of an operating reactor pressure vessel implies good knowledge of the neutron flux at the operating pressure vessel (such knowledge at this time is quite limited) as well as a knowledge of the extent of NDT increase with neutron exposure based upon a comprehensive experimental program. Both of these factors must be studied carefully if experimental data are to be meaningful for reactor operators.

Another factor, low-cycle fatigue, must be considered in relation to brittle fracture, since low-cycle fatigue in steel structures has been shown to cause the growth of cracks. In order to assess the significance of this factor in a nuclear environment, a program has been undertaken to study the fatigue characteristics of irradiated steels. Preliminary results of this study are presented in a separate report (5).

NEUTRON EMBRITTLEMENT OF PRESSURE VESSEL STEELS

The study of neutron embrittlement of steels, as an extension of the NRL brittle fracture study with no direct ties to a specific reactor development project, has covered a range of materials as well as a range of exposure conditions. The primary objectives of the study are the better understanding of the changes in the properties of reactor structural materials and of the engineering significance of these changes. Secondary objectives include the evaluation of irradiation data to suggest mechanisms of embrittlement and possibly to assist in the selection of steels better suited for application in a nuclear environment.

The experimental program has emphasized the steels as well as thermal and nuclear environmental conditions appropriate to the current generation of boiling-water and pressurized water reactors. However, in keeping with the broad nature of the research effort, the program has not been limited to the materials and conditions of current reactors but has included steels of higher strength for potential future nuclear application. The preponderant usage of A212-B and A302-B steels for the pressure vessels of current nuclear power reactors is demonstrated in Table 1 (6). The NRL program has emphasized these two steels along with the A350 steels which were used in Army reactors. The composition and heat treatment as well as the initial NDT values of the several steels studied are presented in Table 2. Unless otherwise noted, these are the steels referred to in subsequent discussion.

Experimental Approach

The experimental approach has involved the irradiation of a large number of Charpy V-notch specimens, and in some cases, drop-weight test specimens as well, in a neutron flux of intermediate intensity so that temperature control was not intolerably complex and yet the irradiation period was not unreasonably long. The most useful reactor facilities utilized have been the core irradiation positions of the Oak Ridge Low Intensity Test Reactor (LITR). Capsule design has emphasized simplicity in order to permit the conduct

Table 1
Power Reactor Pressure Vessel Steels*

Pressure Vessel Steel	Reactor	Thickness of Vessel (in.)
A212-B	Vallecitos ESR	3-1/2
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	2-1/2 (1/4 in. of cladding)
A302-B	Humboldt Bay	4-5
A302-B	Dresden	5-3/8
A302-B	Big Rock Point	5-1/2 (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)

*Compiled from tabulations of DiNunno and Holt (6).

of a large number of irradiations for comprehensive investigation of the effects of the major experimental factors. Neutron dosimetry has been an important part of the experimental program. Dosimeters have been included in all irradiation assemblies. More detailed reports regarding various phases of the experimental approach have been published previously (7-10).

The major experimental variables studied include: the neutron exposure, the type of steel, the irradiation temperature, and the nuclear environment. The effects of varying temperature during irradiation as well as the effects of applying heat treatment cycles for the relief of neutron embrittlement have also been studied. Since each of the major variables is controllable in the design of reactor systems, it has been considered important to determine the effects of each as well as the integrated effects of the several factors.

Effects of Neutron Exposure

Neutron exposure is the most difficult variable to assess in the study and application of steel embrittlement data. Although good progress has been made toward measuring the neutron exposure in a meaningful manner for experimental studies, the lack of similar measurements at the pressure vessel of most nuclear power plants severely limits the prediction of service properties from neutron embrittlement data. The best available compilation of peak pressure vessel exposure levels for sixteen major commercial and military reactors having carbon steel vessels was made by DiNunno and Holt for the U.S. Atomic Energy Commission in 1961 (6). Neutron exposure data from this survey were tabulated by the authors in an earlier report (7). Of the sixteen reactors surveyed, flux

Table 2
Description of Major Steels in Irradiation Study

Item	Steel	Form	Thickness (in.)	Initial NDT (°F)	Chemical Analysis (%)								
					C	Mn	Si	P	S	Ni	Cr	Mo	Other
1	A302-B	Plate	6	+ 10	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	- 30	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	-100	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	-190	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	- 40	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	- 80	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												
7	A336	Forging	11	+ 10	0.19	0.65	0.26	0.011	0.014	0.79	0.40	0.64	0.12 Cu
	Heat treatment: Forged in the temperature range of 1500°F min to 2300°F max. Post-forging treatment: austenitized at 1575°F; water quenched; tempered at 1220°F												
8	17-4 PH	Rod	5/8	- 10*	0.03	0.30	0.60	0.019	0.015	4.24	16.36	-	0.28 Cb
	Heat treatment: Solution treated at 1875 to 1925°F; air cooled; hardened at 1100°F for 4 hours; air cooled												
9	A353	Plate	1	-300*	0.10	0.47	0.29	0.008	0.023	8.99	0.06	0.15	
	Heat treatment: Double normalized 1650°F for 1 hour; air cooled; 1450°F for 1 hour; air cooled; tempered at 1050°F for 1 hour; air cooled												

*Based on Charpy-V 30 ft-lb transition.

values for only three reactors represented direct measurements of the flux in the operating nuclear plant. The calculated and measured values from the survey present a range of peak pressure vessel neutron exposures over the anticipated life of the vessels from 5.6×10^{17} to 2.3×10^{20} n/cm^2 (>1 Mev). Only seven of the reactors surveyed indicated lifetime exposures greater than 1×10^{19} n/cm^2 (>1 Mev). Nevertheless, the exposures expected are sufficient to suggest the need for direct neutron flux measurements at the vessels of several reactors as well as the need for experimental programs to determine the extent of radiation-induced embrittlement under specific reactor conditions.

While no major program for specifically studying the effect of neutron exposure in the absence of other variables has been attempted, many exploratory irradiations have been conducted at temperatures below 450°F, where irradiation temperature effects are small. This body of data permits a general evaluation of neutron exposure effects. A summary trend band developed by NRL studies which relates NDT increase to neutron exposure has been discussed earlier (7, 10). Figure 1 presents the summary trend as extended by more recent data. The boundary lines were established from approximately forty low-temperature irradiations which include eight steels from plates, welds, and forgings. These data represent irradiated properties as determined from drop-weight tests or Charpy V-notch tests using the particular Charpy-V energy level which corresponds to NDT in the unirradiated condition. The consistency of the established trend is

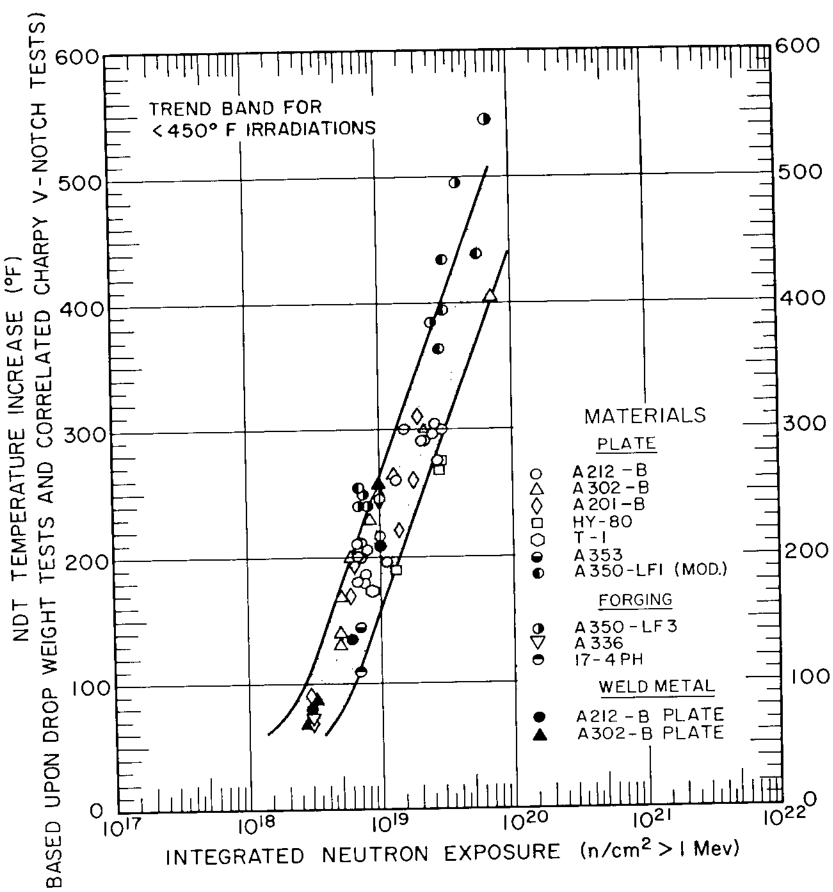

Fig. 1 - Increase in the NDT temperatures of steels resulting from irradiation at temperatures below 450°F

striking when the experimental variables are considered. It should be noted that these data represent irradiations in various positions of several test reactors. When the factors of material and temperature differences are eliminated, the uniformity of the trend of NDT increases with neutron exposure (>1 Mev) is quite vivid. A summary of embrittlement data for a single heat of A302-B steel is presented in Fig. 2. The data represent

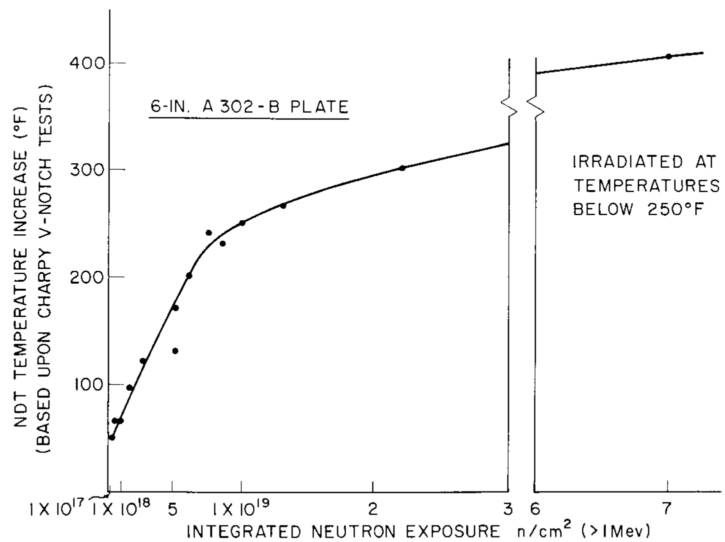

Fig. 2 - Increase in the NDT temperature of A302-B steel irradiated to various integrated neutron exposures at temperatures below 250°F. Linear graph permits direct comparison of effects for various exposure levels.

observed increases in NDT for integrated neutron doses between 3.5×10^{17} and 7.0×10^{19} n/cm^2 (>1 Mev). The irradiation temperature in each case was less than 250°F. The irradiations were conducted in three different reactors; the low dosage points from the Industrial Reactor Laboratory (IRL) Reactor (11), the intermediate dosage values from core experiments in the Oak Ridge Low Intensity Test Reactor (LITR), and the high dosage value (7×10^{19}) from an irradiation in the Materials Test Reactor (MTR). In spite of the fact that all three reactors are light water moderated, neutron spectrum variations are anticipated since the IRL data involved irradiations in a steel block simulating a reactor pressure vessel wall (i.e., several inches removed from nuclear fuel), while the LITR data involved positions near nuclear fuel, and the MTR value involved a position in the beryllium reflector region outside the reactor core. The consistency observed in spite of variations in the nuclear spectrum suggests that the key factors causing the variations in neutron embrittlement demonstrated by the scatter band of Fig. 1 are materials and temperature rather than nuclear environment. A similar graph for low-temperature irradiations of A212-B steel is presented in Fig. 3.

The remarkable consistency of the NDT data when referenced to neutron dosages (>1 Mev) enhances the validity of the >1 Mev criterion for reporting neutron dosage; at least for the case of light water reactors.

If the embrittlement data for other steels are similarly isolated, the trends for individual steels, while not so consistent as that for A302-B, do follow individual trend lines. The trends for several steels have been reported (7). While these trends are generally

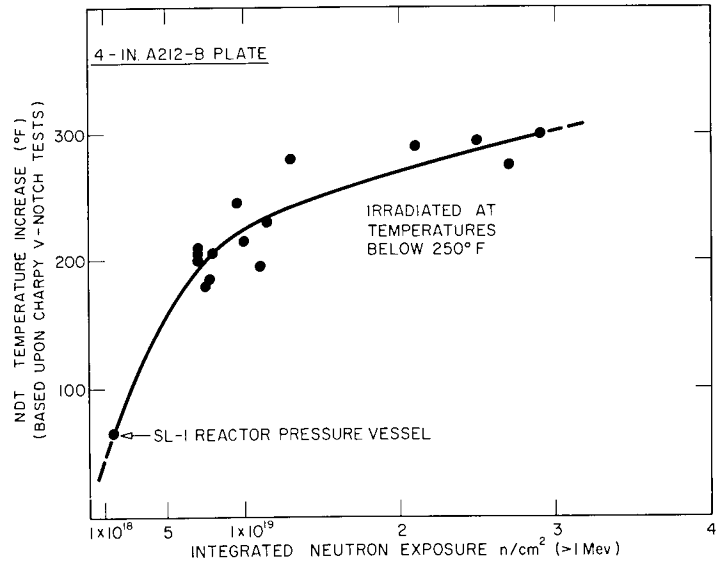

Fig. 3 - Increase in the NDT temperatures of A212-B steel irradiated to various integrated neutron exposures at temperatures below 250° F, SL-1 vessel at 420° F. Linear graph permits direct comparison of effects for various exposure levels.

quite similar, to characterize the pattern ΔNDT versus integrated neutron dose data by a mathematical expression would require a different expression for different steels and for different temperature ranges. Therefore, such expressions are not considered practical except to define a trend or general limit for correlating embrittlement data to reactor operations.

The embrittlement trend for A302-B steel (Fig. 2) shows a very rapid increase in NDT to about 1×10^{19} n/cm^2 with a smaller rate of embrittlement to 7×10^{19}. Complete saturation of radiation embrittlement effects has not yet been observed, however. The constantly decreasing rate of embrittlement has important implications to the problem of embrittlement relief through annealing treatment.

In general, no major discrepancies in experimental observations to date can be related specifically to variations in neutron energy spectrum or to the rate of neutron dosage accumulation. One long-term experiment which was conducted in the Brookhaven Graphite Reactor, however, does show some discrepancy from the summary of ΔNDT data collected in light water moderated reactors (7). The factors of dose rate cannot be separated from energy spectrum in this experiment, however, so the question of possible dose rate and spectrum effects will probably await the evaluation of power reactor surveillance specimens and the determination of the properties of pressure vessels of nuclear reactors no longer in service.

Early surveillance data have been reported (12) and more data of this type will be forthcoming in the near future. The SL-1 reactor vessel has been tested and the availability of other vessels including the Organic Moderated Reactor Experiment and Army PM-2A reactor is anticipated. Data from these sources must be compared to assure the greatest reliability of steel embrittlement data and the most useful application of these data in predicting changes in operating reactor vessels. In this regard, the necessity of careful measurement of the neutron environment in both test reactor experiments and in operating nuclear plants cannot be overstated.

Comparison of Embrittlement Between Steels

The initial compilation of data in terms of ΔNDT as a function of neutron exposure indicated a trend which was relatively independent of steel composition and form. With additional data covering a greater variety of steels and including data from additional irradiation facilities, the basic trend has been broadened as indicated in Fig. 1. One of the key factors to the development of the observed scatter band is the difference in embrittlement response of the steels studied. For example, if the data of Fig. 1 are separated by materials, it is apparent that the band extends from the data for the A350 steels on the left to that for the HY-80 on the right with the mass of data for A212-B and A302-B steels between. The data making up the trend band involved very limited experimentation for the direct comparison of steel response. However, two experiments, one involving irradiation at ~240°F and the other at ~490°F, have been conducted to compare embrittlement response between steels. The 240°F irradiation which included the two grades of the A350 steel, A212-B, A353, and 17-4 PH steels, was an effort to assess the effects of major alloying elements, especially nickel. The neutron exposure for this experiment was 7×10^{18} n/cm^2 (>1 Mev). The pattern of NDT values, which ranged from 225°F for the A350-LF1 to 110°F for the 17-4 PH, does not permit any specific conclusions regarding compositional effects on embrittlement response without further study.

The second experiment included a different group of five steels and involved an exposure of 1.4×10^{19} n/cm^2. The results of the 490°F irradiation are presented in Table 3 along with data for the same steels which were irradiated in the 240°F experiment.

Table 3
Increase in NDT Temperature of Irradiated Steels

Pressure Vessel Steel	Initial NDT	ΔNDT (°F) After 490°F Exposure to 1.4×10^{19} n/cm^2 (>1 Mev)	ΔNDT (°F) After 240°F Exposure to 7.0×10^{18} n/cm^2 (>1 Mev)
A350-LF3	- 80	270	240
A212-B	- 30	230	200
A353	-300*	200	145
A302-B	+ 10	200	-
HY-80	-190	100	-

*Based upon 30 ft-lb Charpy V-notch transition.

With reference to the data in Table 3, the quenched and tempered HY-80 steel shows the smallest change, while the A350 steel showed the greatest change; the difference between their shifts was greater than a factor of 2. The more commonly used A212-B and A302-B steels fall in the high to midrange of ΔNDT values. For reference purposes, the data of Table 3 are presented in Fig. 4 over the NRL ΔNDT trend band. The spread of data suggests that the specific steel used in a reactor pressure vessel may be an important factor in assessing the embrittlement effects of neutron exposure for that reactor.

Another experiment involving laboratory heats of steels with uranium additions also demonstrated differences in embrittlement response between steels. In this experiment, the notch ductility properties of three heats of steels containing various additions of natural uranium and a commercial heat, each prepared to the nominal composition of A212-B, were compared to determine the effects of the uranium additions. Charpy

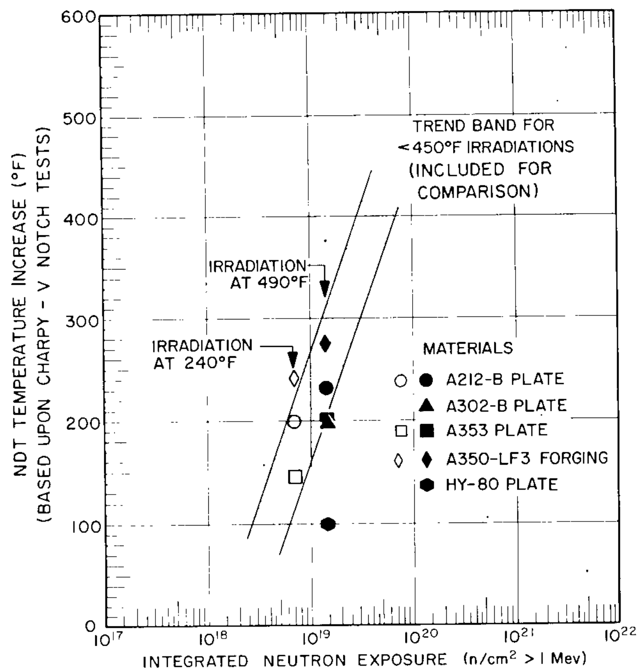

Fig. 4 - Increases in the NDT temperatures of steels irradiated simultaneously in two experiments conducted respectively at temperatures of 240° and 490°F

V-notch specimens of the four steels were irradiated simultaneously at ~240°F to a neutron dosage of 1.3 x 10^{19} n/cm^2 (>1 Mev). Comparisons of the notch ductility behavior of uranium-bearing laboratory heats to that of the reference commercial heat are given in Table 4 and in Fig. 5. The results of postirradiation annealing which will be discussed in a later section are also shown in Fig. 5.

In summary, the 0.06%-uranium steel appeared superior in terms of notch ductility, to both the uranium-free and 1.1%-uranium steels and to the 4-in. plate of a large

Table 4
The Effect of Irradiation on the Notch Ductility Properties of Uranium-Bearing Steels

Material	Plate Thickness (in.)	30 ft-lb Transition (°F)			Energy Absorption at Full Shear (ft-lbs)		
		Unirradiated	Irradiated	ΔNDT	Unirradiated	Irradiated	Δft-lbs
0% U	1/2	- 30	145	175	72	64	- 8
0.06% U	1/2	-105	5	110	110	94	-16
1.1% U	1/2	140	285	145	37	72	+35
Commercial Heat	4	5	285	280	82	46	-36

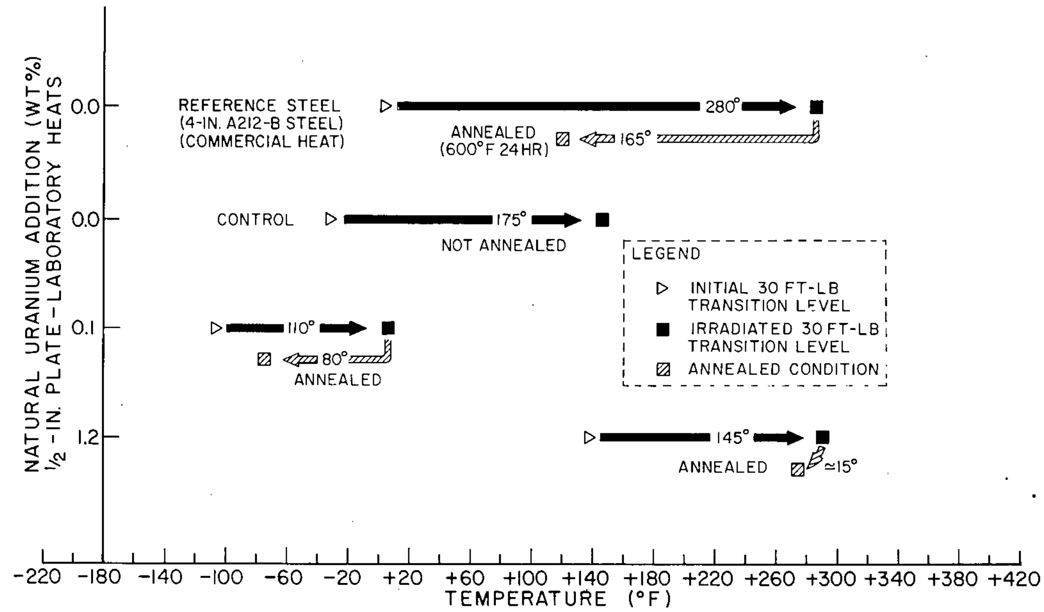

Fig. 5 - 30 ft-lb Charpy-V transition of uranium-bearing steels before and after irradiation at 240°F to 1.3×10^{19} n/cm^2 > 1 Mev. Results of postirradiation annealing are also shown.

commercial steel heat. Furthermore, in comparing the results of the uranium-free laboratory steel and the 4-in. commercial steel, a significant difference in the radiation effect was observed. This suggests that steelmaking practice may be an important factor in the development of steels which are less sensitive to nuclear radiation.

Further studies are being conducted in order to explain the differences in radiation sensitivity observed in the experiments described above. These studies will involve a series of experiments to assess the effects of chemical composition and metallurgical microstructure.

Another aspect of the variation in embrittlement between steels involves observed differences between heats of the same steel having the same nominal composition. Such heat-to-heat variations have been reported by Carpenter, Knopf, and Byron of the Bettis Atomic Power Laboratory (13). Irradiation experiments including several heats of A302-B steel were conducted by Bettis so that the observed differences between heats could not be attributed to differences in the thermal or nuclear environmental conditions during irradiation. The results presented show that certain heats (designated "sensitive") produced increases in transition temperature which fall in the left portion of the NRL trend band, whereas the heats called "insensitive" fall to the right of the <450°F band. It should be noted that the irradiation temperatures in the Bettis studies were 450 (for most irradiations), 470, and 470 to 520°F, so there exists the possibility for some correction of embrittlement because of irradiation temperature. Nevertheless, the differences observed between heats irradiated in the same capsule are quite significant and can only be considered as results of differences in chemistry or metallurgical microstructure. This observation is reinforced by some recent data obtained at NRL upon materials from two different heats of A302-B steel which were irradiated in a single surveillance capsule in a power reactor. In this case, the steels represented a commercial heat which has been distributed to several laboratories for irradiation evaluation as well as specimens from a heat which was used to fabricate the pressure vessel of the commercial reactor. While the neutron exposure has not yet been determined, it is known to have been very similar for both steels. The irradiation temperature was not measured but was known to exceed somewhat the operating coolant temperature (~515°F) during much of the core life exposure period. The factors of total neutron exposure and irradiation temperature are only important for comparing the surveillance results to other experimental observations. Of more importance is the fact that, while the reference steel showed a transition temperature increase of only 225°F, the pressure vessel steel exhibited an increase of 320°F after irradiation under the same conditions. Thus, the NDT increase for the pressure vessel steel was approximately 50% greater than that of the reference steel which was used in constructing the NRL trend band (Fig. 1). This comparison is based upon an initial energy to fracture of 30 ft-lb at +10°F (nominal NDT) for the pressure vessel steel and +15°F for the reference steel. The actual NDT of the reference steel as determined by drop-weight tests was +10°F.

Implicit in both the Bettis and NRL studies of variations in embrittlement sensitivity between steels is the recurring suggestion that an answer can be found for minimizing steel embrittlement under high-energy nuclear radiation. Additional research for exploring this phenomenon is underway. The ultimate goal involves developing knowledge which will permit the production of steels which have low initial NDT values and consistently show minimum radiation embrittlement.

Effect of Irradiation Temperature

Irradiation temperature is an important factor in establishing the extent of neutron embrittlement, particularly in the temperature range which represents the operating conditions of current power reactors. For irradiation temperatures below 450°F, however, no appreciable temperature effect is observed. This is demonstrated for A212-B and A302-B steels in Table 5. For both steels, irradiation at 550°F is shown to significantly reduce the embrittlement as measured by the ΔNDT value. Similar patterns have been observed for other steels. The reduction in embrittlement above 450°F through a thermal correction process is believed to be the result of enhanced movement of neutron-induced defects which, in effect, restores some of the initial ductility.

The smaller embrittlement at elevated temperatures is demonstrated by Fig. 6 for irradiation temperatures between 500 and 750°F. These data are presented along

Table 5
NDT Increase for Steels Irradiated at Various Temperatures

Steel	Irradiation Temp. (°F)	Neutron Dosage (n/cm^2 >1 Mev)	ΔNDT (°F)
A212-B (4-in. 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-in. 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

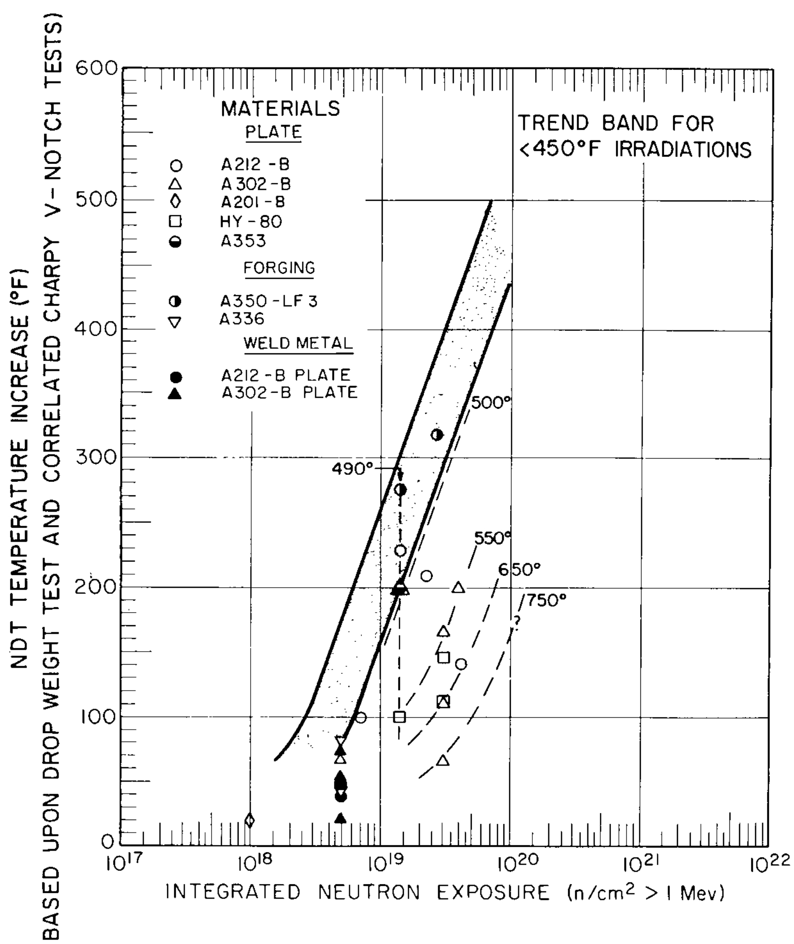

Fig. 6 - Increase in the NDT temperatures of steels resulting from irradiation at temperatures above 450°F. Points at 5 x 10^{18} n/cm^2 represent early irradiations in the Brookhaven Graphite Reactor at 500° to 600°F.

suggested trend lines for various temperatures with reference to the trend band for irradiations at temperatures at or below 450°F. No significant departure from the slope established for low-temperature irradiations is evident. The progressive and appreciable reduction in embrittlement resulting from irradiation at higher temperatures is apparent. This factor may be of great significance in assessing the effects of high-energy neutron irradiation on the structural components of future reactors.

Variations which may be related to the materials differences are not readily apparent in Fig. 6, although the separation of data points on the 500°F trend line, the low point for HY-80 irradiated at 490°F, and the lone point for the A350-LF3 (uppermost point, involving irradiation at 510°F), suggest compositional or microstructural effects. However, evaluation of the significance of these differences in materials will depend upon additional determinations.

Another experiment, involving the simultaneous irradiation of A302-B and HY-80 steels at three different temperatures, 540, 640, and 740°F, has provided some valuable data for developing trends in elevated temperature embrittlement. The total neutron exposure was 3.1 x 10^{19} n/cm^2 (>1 Mev). The NDT increases for the A302-B were 165, 110, and 65°F, respectively, after irradiation at 540, 640, and 740°F. For the HY-80, the increases were 145, 110, and 225°F, the last showing a departure from all previous observations. This anomaly is thought to be the result of the combined effects of neutron embrittlement and neutron enhanced temper embrittlement.

Effects of Varying Temperature During Irradiation

The design of most boiling and pressurized water reactors permits operation at any one of several temperatures over a range of 100°F or more. Although most nuclear power plants operate so that the temperature of the pressure vessel is rather consistent, variations may occur. For example, the design temperature of the Dresden reactor is 621°F while the normal operating temperature is 545°F. The design temperature of the SM-1A is 600°F but it normally operates at 430°F. Since there is a possibility for the pressure vessel temperature to be either raised or lowered, two experiments were conducted to test the results of changing the exposure temperature during irradiation. In one case, the temperature was raised for the latter part of the irradiation, while in the other experiment the temperature was lowered for the last portion of the exposure period.

Effects of Low-Temperature Irradiation Followed by Continued Exposure at Elevated Temperature — In order to explore the effects of low, then high, irradiation temperatures, an instrumented assembly containing specimens of A302-B steel was irradiated according to a two-phase schedule. Phase one entailed exposure at 400°F. The transition temperature shift developed during this period was estimated from the results of earlier irradiations of this steel at <450°F in the same reactor facility. Immediately following phase one, the irradiation was continued but at a higher temperature, 540°F, which is known to produce thermal annealing during exposure. The estimated exposure during phase one was 2.3 x 10^{19} n/cm^2 with an added 1.6 x 10^{19} n/cm^2 during phase two. Comparison of the notch ductility behavior of the A302-B steel at the termination of each exposure phase (Fig. 7) indicates that considerable annealing of the estimated phase one embrittlement occurred during the second phase exposure at 540°F. It should be noted that the net increase in NDT after the second phase irradiation is about 100°F less than that estimated after the first phase in spite of a total neutron exposure which is 170% of that of the first phase. One point from a previous irradiation of A302-B at 540°F is also shown in Fig. 7 (open circle). The close agreement between the two 540°F irradiation experiments suggests that the ultimate behavior of this steel was not affected by the low temperature of exposure during phase one and that the total shift in transition temperature in this case is determined by the final irradiation temperature in conjunction with the total neutron exposure received. A similar response was noted for A350-LF3 steel in a comparable experiment. These data may have direct significance for nuclear power reactors. For example, if a pre-established, maximum allowable NDT is being approached in a power reactor vessel, an increase in operating temperature may lower the existing NDT temperature and thus extend service life. This premise, however, is necessarily based upon the assumption of a relatively large increase in operating temperature in order to produce a significant degree of recovery. Furthermore, engineering considerations for such a procedure would require careful attention to the possible effects of higher stresses in the vessel during operation at the higher temperature.

Effects of Elevated Temperature Irradiation Followed by Continued Exposure at a Lower Temperature — In order to test the effects upon steel embrittlement of a reduction of temperature during irradiation, a two-phase experiment was conducted in which five steels were subjected to irradiation at 490°F to an integrated neutron exposure of 1 x 10^{19} n/cm^2 with an added exposure of 5 x 10^{18} n/cm^2 at 350°F. This experiment was conducted in conjunction with the 490°F irradiation of five steels which was presented above (Table 3 and Fig. 4). The comparative results of the single-phase irradiation and the

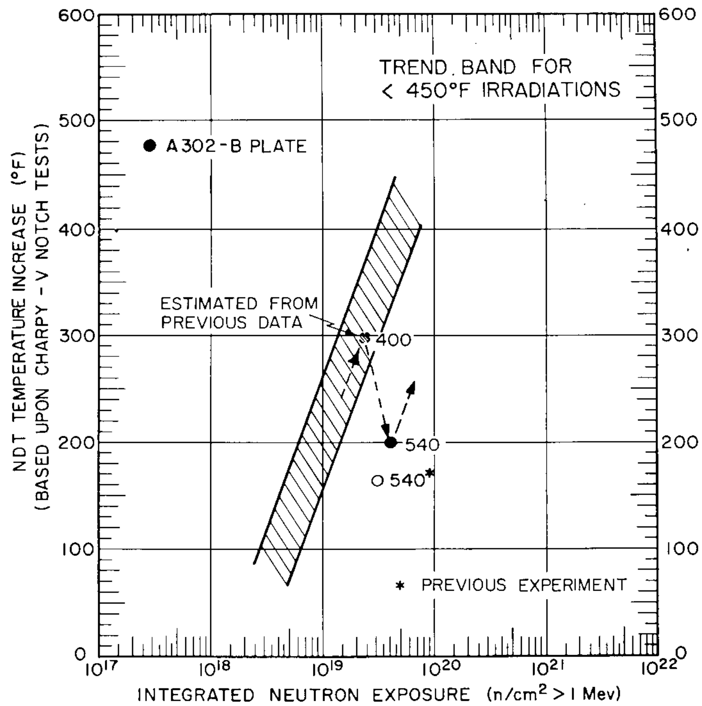

Fig. 7 - NDT temperatures of A302-B steel resulting from irradiation in a two-phase (two temperature) schedule; first at low temperature (400°F) and then at a higher temperature (540°F)

two-phase irradiation are presented in Table 6. The small differences in total neutron dosage received (1.4×10^{19} n/cm^2 vs 1.5×10^{19} n/cm^2) by the two material conditions arose from the small neutron flux gradient over the length of the experimental unit. As expected, the two-phase exposure resulted in a greater transition temperature increase as compared to the referenced condition, but the magnitude of the increase was not the

Table 6
Notch Ductility Behavior of Five Steels Irradiated Continuously at 490°F Versus Irradiation in Two Phases*

Material	ΔT (°F) 1.4×10^{19} n/cm^2 at 490°F	ΔT (°F) 1.0×10^{19} n/cm^2 at 490°F plus 5×10^{18} n/cm^2 at 350°F	Difference ΔT (°F)
A212-B	230	255	25
A350-LF3	270	335	65
A353	200	240	40
A302-B	200	230	30
HY-80	100	170	70

*490°F followed by a period at 350°F.

same for all five steels. However, in spite of the observed differences in annealing characteristics, the order of irradiation response remains generally consistent with previously observed trends except that the HY-80 showed a greater increase with the low-temperature irradiation. Nevertheless, the order of net change in NDT is the same as that previously observed.

These observations, when applied to nuclear plants, suggest that to lower operating temperatures invites greater embrittlement of steel pressure vessels and thus should be avoided if at all possible.

EMBRITTLEMENT RELIEF THROUGH ANNEALING TREATMENT

With NDT increases as great as 545°F (Fig. 1) and the severe reduction in energy to fracture as shown in Figs. 8 and 9, the development of some means for the restoration of notch ductility becomes important. The need for embrittlement relief is especially important for steels irradiated at temperatures below 450°F. The application of heat treatment cycles was explored for this purpose.

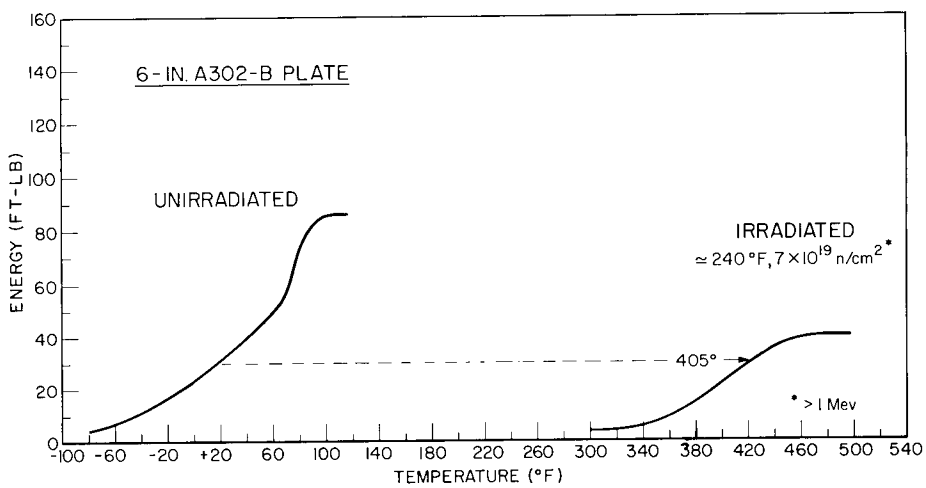

Fig. 8 - Transition temperature and shear energy characteristics of irradiated A302-B steel

NRL studies of annealing response have been directed toward an evaluation of pertinent variables: the material, the irradiation temperature, the neutron exposure, and the duration and temperature of annealing. These studies have evolved in three stages, beginning with a single irradiation and annealing treatment through an intermediate stage of cyclic irradiation and annealing to the direct simulation of in-service annealing. Data from these experiments may be of direct engineering interest to any decision to anneal the pressure vessel of an operating nuclear power plant.

Results of Single Postirradiation Annealing Treatment

The first phase of the investigations on annealing behavior involved irradiation of steels at a particular temperature followed by heat treatment under selected conditions. Single-phase annealing treatments revealed differences in recovery response between

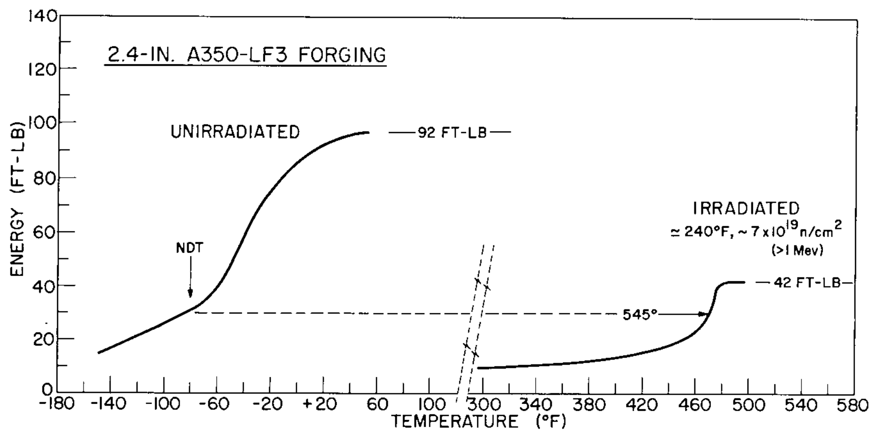

Fig. 9 - Transition temperature and shear energy characteristics of irradiated A350-LF3 steel

steels as well as variable degrees of recovery when different annealing temperatures and durations were utilized. The variable response of several steels to the same annealing treatment after a particular irradiation has been reported (7). In brief, five steels which were irradiated at 240°F developed a spread of ΔNDT values from 110 to 255°F. After annealing for 168 hr at 600°F, the recovery pattern showed the smallest recovery for the steel which demonstrated the smallest increase in NDT and greatest recovery for the steel showing greatest ΔNDT. No explanation for this behavior has yet been determined.

Another experiment which emphasized the materials factor involved the annealing of the irradiated uranium-bearing steels (Fig. 5). In this case, annealing at 600°F for 24 hr resulted in an anomalous pattern of results (Table 7). The recovery pattern in this case was contrary to that recorded above in that the 0.06%-uranium steel which showed the least embrittlement showed the greatest percentage annealing recovery. This anomalous behavior is not understood and requires additional study for clarification.

The effects of annealing after different integrated neutron exposures have not been investigated in connection with single-phase irradiation-annealing procedures. However,

Table 7
The Effect of Postirradiation Annealing* on the Notch Ductility Properties of Uranium-Bearing Steels

Material†	Plate Thickness (in.)	30 ft-lb Transition			% Recovery
		Initial (°F)	As Irrad. ΔT (°F)	Irrad. plus Anneal ΔT (°F) Recovery	
0.06% U	1/2	-105	110	80	73
1.1% U	1/2	140	145	15	10
Commercial Heat	4	- 30	280	165	59

*600°F - 24 hr.
†0% - U steel not annealed.

the effect of irradiation temperature on subsequent annealing response has been investigated and generally shows that greater annealing recovery, in terms of percentage of ΔNDT, is possible after irradiation at low temperatures. The possibilities for embrittlement relief are progressively smaller with successively higher irradiation temperatures if the same annealing temperature is used. For example, specimens of A212-B steel were irradiated simultaneously at 275 and 510°F, respectively. Both groups received the same neutron dosage but the 275°F group exhibited recovery of 85.6% after annealing at 750°F for 36 hrs. The recovery for the 510°F group was 64%. Similar observations have been made for other steels and for other temperature ranges. On the other hand, if steels are irradiated at different elevated temperatures (such as 500, 600, and 700°F) and a given temperature increment (such as 150°F) is added for annealing, there appears to be greater recovery for the higher irradiation temperature. For example, two groups of A302-B specimens were irradiated simultaneously to a dosage of 3×10^{19} n/cm^2 at 640 and 740°F, respectively. The 640°F group which was annealed at 800°F, showed a recovery of 64% vs an 85% recovery for the group irradiated at 740°F which was subsequently annealed at 900°F. Thus, current data suggest that, in addition to smaller initial embrittlement following irradiation at elevated temperatures, the possibilities for annealing recovery are improved by elevated temperature irradiation.

The increasing effectiveness of annealing with longer times and higher temperatures has been observed as anticipated. This trend is demonstrated in Fig. 10 which presents the results of the irradiation of A350-LF1 steel at 430°F followed by annealing at three temperatures for various time periods. The extent of recovery even with an annealing temperature of 600°F showed considerable promise for correcting radiation embrittlement. However, the promise was tempered by the lack of knowledge of what might happen with multicycle, irradiation-annealing treatments.

Cyclic Irradiation-Annealing Treatment

Results of multi-cycle experiments on A350-LF3 and A212-B steels involving irradiation at <240°F and annealing at 700°F have been presented (7). These experiments suggested that the rate of embrittlement was essentially the same after a full irradiation-annealing cycle as with the virgin material. Nevertheless, in spite of a cumulative increase in the NDT with successive cycles, the projected NDT increase with intermediate annealing is considerably less than the NDT without annealing. Thus, some benefit for irradiation-annealing treatment cycles is indicated. The observed benefits were greater for the A350-LF3 steel than for the A212-B.

A similar cyclic experiment involving A302-B and HY-80 steels was also conducted. The two steels were irradiated at $\sim$240°F with intermediate annealing at 650°F for 24 hrs. The notch ductility behavior of these steels after various stages of two complete irradiation-annealing cycles with reference to the NRL trend band of Fig. 1 is shown in Figs. 11 and 12. In both these figures, the shaded triangles represent extrapolated or estimated values and suggest that, for the A302-B, there is a measurable advantage for the annealed condition, while the accumulated ΔNDT for the HY-80 steel with annealing is almost the same as that without annealing.

While the extent of embrittlement relief with cyclic irradiation and annealing was not great in terms of percentage reduction, the net embrittlement relief may be important when considered in terms of accumulative exposure to the reactor vessel. Accordingly, experiments for simulating periodic annealing treatment of operating reactor vessels were undertaken.

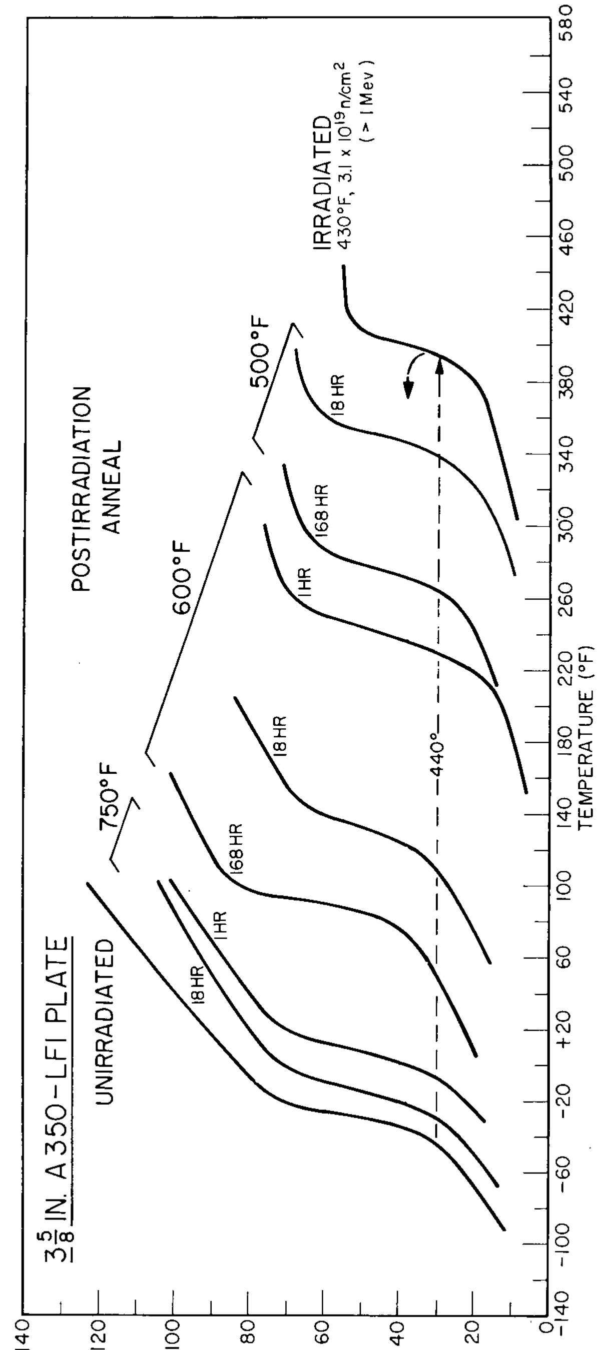

Fig. 10 - Notch ductility characteristics of irradiated A350-LF1 steel. Results of postirradiation annealing show effects of various heat treatment (time-temperature) combinations.

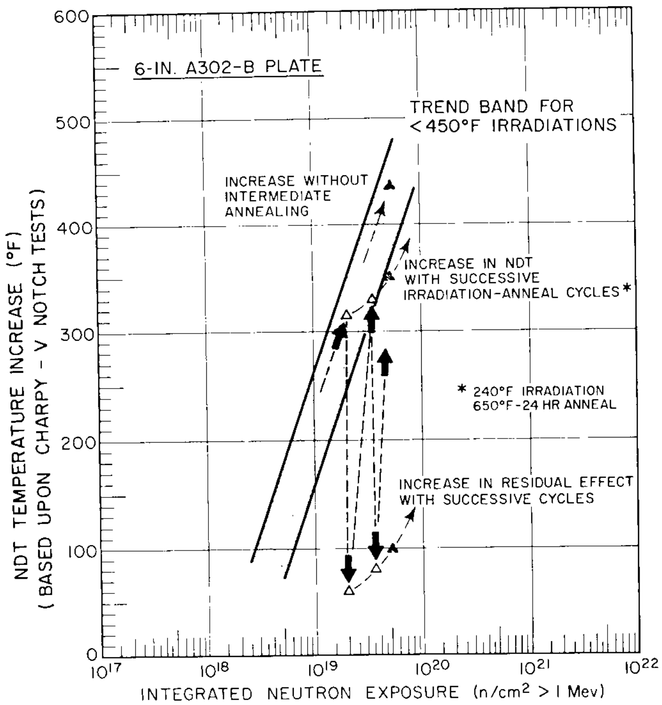

Fig. 11 - NDT temperature behavior exhibited by A302-B steel at various stages of cyclic irradiation-annealing treatments

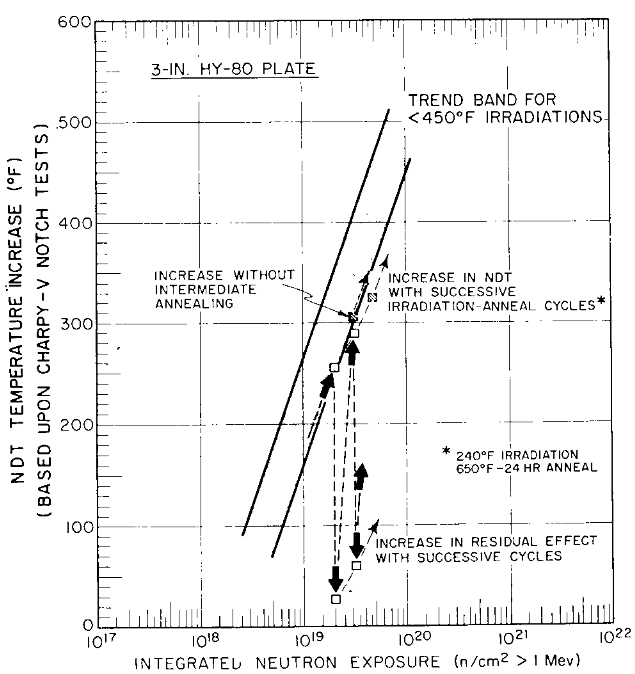

Fig. 12 - NDT temperature behavior exhibited by HY-80 steel at various stages of cyclic irradiation-annealing treatments

Simulation of Periodic In-Service Annealing

The ultimate assessment of heat treatment for embrittlement relief involves a direct evaluation of in-reactor irradiation with intermediate annealing cycles. One experiment of this nature has been conducted using the A350-LF1 steel. This experiment assumed a pressure vessel temperature of 430°F and an annealing temperature which was within that attainable in a pressurized water reactor without external heat sources. The plan of the experiment, which contained four separate groups of Charpy V-notch impact specimens, is outlined diagrammatically in Fig. 13. The experiment was intended to determine the effect of annealing frequency and the effects of two different annealing temperatures, 550 and 600°F. One section was annealed three times in-reactor and the other two only once. Second and fourth annealing treatments were conducted out-of-reactor after the fourth irradiation cycle. The four groups of Charpy specimens received slightly different neutron exposures ranging from 2.8×10^{19} n/cm² (>1 Mev) in the control (constant temperature) section to 3.6×10^{19} n/cm² in the top section. The estimated ΔNDT values associated with these neutron exposures are shown in Table 8. It is readily apparent that, under these experimental conditions, the frequency of annealing is important to net improvement in notch ductility. The single, in-reactor annealing cycle provided so little recovery that it was considered of no practical value. Even the recovery observed with three intermediate annealing cycles appears small. However, when it is considered with reference to the trend of NDT increase with neutron exposure, a different conclusion may be reached. Figure 14 shows the relative positions on the ΔNDT trend line for low temperature (<450°F) irradiation of the A350-LF1 steel for the two conditions, with and without intermediate annealing treatment. The estimated ΔNDT value of 455°F at 3.6×10^{19} n/cm² (>1 Mev) is compared with a 315°F actual ΔNDT which is equivalent to only 1.3×10^{19} n/cm². Thus, the intermediate annealing treatment under these particular experimental conditions may be considered to have corrected the embrittlement created by an increment of exposure equal to $\sim 2.3 \times 10^{19}$ n/cm², almost two-thirds of the total exposure.

Applying this to an operating reactor, it appears the intermediate annealing, in some cases, might be useful for reducing embrittlement. Of course, a net reduction of 50% or more is highly desirable. An experiment now underway will test the effects of irradiation at 430°F with intermediate annealing at higher temperatures (650 to 750°F). Another

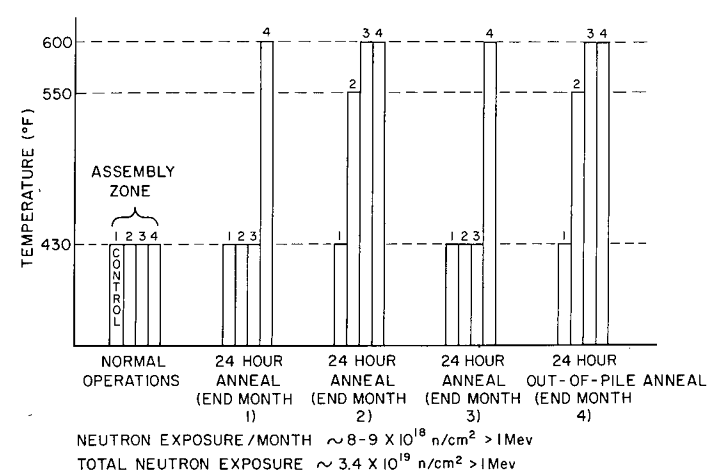

Fig. 13 - Schedule for the periodic in-reactor annealing of A350-LF1 steel during irradiation at 430°F

Table 8
Effect of Intermediate Annealing on the NDT Increase of Irradiated A350-LF1 Steel

Material Condition	Neutron Exposure (x 10^{19} n/cm^2 > 1 Mev)	Measured ΔNDT (°F) With Annealing	Estimated* ΔNDT (°F) Without Annealing	Net Gain With Annealing	
				ΔT (°F)	(%)
Control	2.8	415	-	-	-
Annealed mid-cycle 550°F, 24-hr	3.1	405	430	25	5.8
Annealed mid-cycle 600°F, 24-hr	3.3	400	440	40	9.1
Annealed quarter-cycle 600°F, 24-hr(3X)	3.6	315	455	140	30.8

*Based on previous 430°F irradiations of this material.

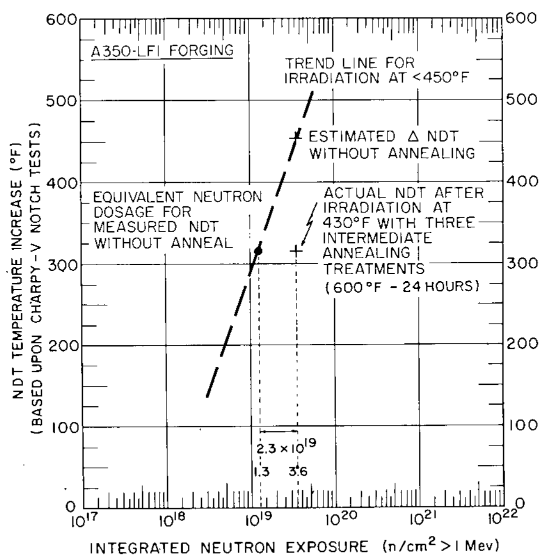

Fig. 14 - Increase in NDT temperature for A350-LF1 steel with intermediate annealing treatment. Reference to NDT trend line indicates extent of correction resulting from the annealing treatment. Correction of 140°F represents neutron exposure of 2.3 x 10^{19} n/cm^2.

current experiment involves the irradiation of A212-B and A302-B steels at 550°F with intermediate annealing at 650, 750, and 800°F for 48 hrs. Preliminary results of this experiment indicate that the net ΔNDT with intermediate annealing at 750°F (twice) and 800°F (once) is only about 60% of the ΔNDT observed for the 550°F irradiation with no annealing treatment. Thus, the value of this technique may be quite important for those reactors in which higher annealing temperatures may be utilized for a period of days.

RELATING EXPERIMENTAL OBSERVATIONS TO NUCLEAR POWER REACTOR CONDITIONS

Most of the results reviewed above were obtained in accelerated test reactor irradiation experiments in which efforts have been made to simulate power reactor conditions. While the data reported cover a broad range of materials and environmental conditions, several questions regarding the direct applicability of the experimental data remain. These questions include: the effects of applied stress during irradiation, the effects of slow vs fast rates of neutron dosage accumulation, the effects of different neutron energy spectrum conditions and the variable factors associated with the pressure vessel as fabricated. Long-term surveillance experiments and the direct evaluation of pressure vessels removed from nuclear service are the two means now available for correlation of data from test reactor experiments with the operating nuclear condition.

Radiation damage surveillance programs which have been devised for several commercial nuclear power plants were described by DiNunno and Holt (6). Initial data from the surveillance of the Dresden reactor have been reported (12). These data indicate no significant departure from the NRL steel embrittlement data. Preliminary results from another commercial reactor surveillance program have been obtained at NRL. Results of this effort* demonstrate the value of including in each surveillance capsule a well documented reference steel of the same type as the component being monitored, for in this case, a significantly greater embrittlement of the pressure vessel steel was observed. Furthermore, the simultaneous irradiation of a reference steel with the component material may provide a basis for comparative extrapolation of the data.

Surveillance of the Army SM-1A reactor is also being conducted by NRL. Initial surveillance involved the exposure of neutron dosimeter wires along the inside of the pressure vessel as well as steel specimens above the core. Results of the dosimetry surveillance have been determined and shown to agree with critical facility mockup values, thus developing confidence for the extrapolation of dosimetry data for predicting pressure vessel embrittlement. The initial group of surveillance specimens were recently removed from the SM-1A reactor and will be tested in the near future.

With more extensive correlation, neutron dosimetry data based upon the reaction Fe^{54} (n,p) Mn^{54} may be used directly to predict changes to the pressure vessel. 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, in the postaccident evaluation of the SL-1 reactor pressure vessel, the Mn^{54} constituent 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 mandatory that both specimens and neutron dosimeters be used in power reactor surveillance programs so that a direct correlation with other experimentally observed data can be obtained. (Procedures for conducting radiation damage surveillance programs have been suggested by the ASTM in Recommended Practice 185 61 T.)

A possibility for supplementing the results of experimental programs and reactor surveillance programs is presented by reactors which have been removed from nuclear service. Results from the evaluation of the SL-1 reactor pressure vessel have been presented (14). No significant departure from previous NRL experimental observations was observed. In fact, a direct correlation with other NRL data has been shown (11).

Two other nuclear power plants, the Organic Moderated Reactor Experiment (OMRE) and the Army PM-2A reactor have been terminally shutdown and may offer excellent

*See section on Comparison of Embrittlement Between Steels.

opportunities for further evaluation of materials after nuclear service. The pressure vessel of the OMRE has been sampled and neutron dosimetry analysis has been made (15). This survey resulted in a recommendation to the U.S. Atomic Energy Commission that further evaluation be made of the OMRE pressure vessel. Similarly, the PM-2A reactor vessel is to be sampled to assess the extent of neutron exposure and also may be sectioned for further evaluation of the vessel properties.

The development of data from reactor surveillance programs and from materials of reactor components taken out of service will, if in agreement with the accelerated irradiation data, enhance the direct applicability of all data for predicting the embrittlement of reactor pressure vessels. This knowledge will permit reactor operators and reactor designers to take the necessary action for minimizing the possibilities of a brittle fracture of any critical steel reactor component.

SUMMARY AND CONCLUSIONS

This report was prepared to present the latest information regarding neutron embrittlement of reactor pressure vessel steels. Certain background data have also been reported in order to present the new data in proper perspective.

In the course of the NRL investigations of neutron embrittlement, a variety of steels of current and potential future reactor application have been studied under a variety of thermal and nuclear environmental conditions. The standard for determining embrittlement was the nil ductility transition (NDT) temperature as determined directly by drop-weight tests in some cases, but normally by Charpy V-notch specimens correlated to the drop-weight NDT. The engineering significance of the ΔNDT data may thus be applied more broadly using the fracture analysis and fracture-safe engineering concepts which have been developed at NRL with the NDT as the common reference standard.

Certain conclusions as well as certain anomalous observations are presented to summarize experimental results to date.

1. The extent of pressure vessel steel embrittlement has been shown to depend upon the total neutron exposure, the type of steel, and the temperature during irradiation.

(a) Neutron Exposure – A rather consistent trend band for NDT increase (extending to 545°F) with integrated neutron exposure has been observed for several irradiated steels. However, the rate of embrittlement decreases with each added increment of neutron dosage. If the material and temperature factors are isolated, a uniform relationship between ΔNDT and neutron exposure ($n/cm^2 > 1$ Mev) is observed even though various reactor facilities representing wide differences in irradiation rate and possibly in neutron energy spectrum are used. Data from reactor surveillance programs from the SL-1 pressure vessel evaluation and from an experiment simulating a pressure vessel wall have correlated well with other NRL experimental data. Thus, there is no significant 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. In addition, good correlation of notch ductility and dosimetry data for a variety of reactor facilities validates the neutron dosimetry techniques used.

(b) The Type of Steel – Although a trend band has been established which outlines the general embrittlement of carbon and low-alloy steels with neutron exposure, differences in sensitivity have been noted between different steels. Of the steels studied, the A350 steels show the greatest and the HY-80 the least embrittlement. Significant differences in embrittlement have also been noted for heats of the same nominal type of steel. A laboratory heat of A212-B steel when irradiated along with specimens from a commercial heat of the same nominal type, exhibited only two-thirds the embrittlement of the latter. Similarly, a reference heat of A302-B steel exposed along with specimens of an A302-B reactor pressure vessel in a commercial reactor surveillance program showed only two-thirds the NDT increase of the pressure vessel specimens. Thus, knowledge of the irradiated characteristics of a specific pressure vessel steel is important if the irradiated properties are to be predicted for the assessment of operational limitations. The exact cause of these variations, whether compositional or microstructural, or both, is not yet known.

(c) Irradiation Temperature – No significant effect of different irradiation temperatures has been observed for the range 200 to 450°F, although there is evidence of slightly enhanced embrittlement of some steels at ~300°F and at ~450°F. The magnitude of the observed enhancement is not sufficient to be of concern to practical nuclear engineering but may be of interest in the study of mechanisms of radiation embrittlement. When progressively higher irradiation temperatures in the range of 450 to 750°F are employed, progressively smaller increases in NDT are observed. 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 observation indicates the necessity for determining the effects of neutron irradiation on each steel which is considered for nuclear application at elevated temperatures.

Although some differences related to the type of steel have been observed with elevated temperature irradiation, the trends of embrittlement with neutron exposure parallel the data trend for lower (< 450°F) irradiation temperatures.

If the irradiation temperature is lowered during a portion of the irradiation period, accelerated embrittlement takes place. On the other hand, if the temperature is raised during the latter portion of an irradiation cycle, the net increase in NDT for the total irradiation is equivalent to that expected if the higher temperature had been applied during the entire irradiation period.

2. Studies of annealing treatment for embrittlement relief of irradiated steels have shown effects also related to the neutron exposure, the type of steel, and the irradiation temperature. However, the temperature and duration of annealing are the most important factors in annealing results. In general, the annealing studies have demonstrated that steels which exhibit the greatest embrittlement are most readily relieved of embrittlement by heat treatment, that greater percentage recovery is attained for higher irradiation temperatures if the annealing temperatures applied are a given increment (such as 150°F) above the irradiation temperature, and that progressively greater embrittlement relief is attained with longer annealing periods and with higher annealing temperatures.

The results of multicycle irradiation-annealing treatments simulating reactor service conditions are of most direct interest. Exploratory studies indicate that frequent annealing (limited neutron exposure before annealing or between annealing treatments) is beneficial. Net recovery for an exploratory annealing experiment involving the A350-LF1 steel and simulating reactor operation at 430°F (four irradiation cycles) with periodic annealing (three intermediate cycles) at 600°F was approximately 30% of the ΔNDT. For the specific exposure circumstances, the 30% ΔNDT represents about 60% of the neutron dosage. That is, the embrittlement resulting from the latter 60% of the neutron exposure may be considered nullified by the intermediate annealing treatment. On this basis, the annealing treatment may be quite beneficial; however, the benefits must be assessed in terms of a particular reactor situation. Preliminary results of additional irradiation-annealing experiments involving other steels (A212-B and A302-B) and higher temperatures are very encouraging. This investigation is continuing.

3. Specimens from various reactor surveillance programs and from deactivated reactor pressure vessels (such as those from the PM-2A and OMRE reactors) should be evaluated and compared with data from accelerated experimental programs in the near future. Such a comparison will add greater engineering significance to the body of experimental data now available and should satisfactorily answer questions as to the effects of applied stress and possible differences in steel embrittlement resulting from variable nuclear conditions.

In applying the results of irradiation embrittlement studies to the problem of potential pressure vessel fracture, there is a natural and proper tendency toward conservatism. Nevertheless, several factors favoring optimism toward overcoming this problem in the future may be suggested in summary. In addition to the general conservatism in nuclear pressure vessel design and the emphasis upon design and quality control for minimizing stress concentrations and flaw inclusions, there are several bright spots in the experimental embrittlement data. Some of these favorable factors are outlined with reference to limiting observations as well.

(a) For most commercial reactor pressure vessels, the neutron exposures required for significant increases in NDT (several hundred degrees Fahrenheit) are not anticipated during the life of the vessel. However, for a particular reactor, the effect of any changes such as power level or core loading variations which might raise anticipated pressure vessel exposures must be considered before the problem is dismissed.

(b) The observed differences in embrittlement "sensitivity" between steels suggest that with more complete experimental knowledge, a better understanding of the mechanisms of neutron embrittlement will be possible and this, in turn, should make possible the production and application of steels which are relatively insensitive to radiation. A favorable aspect of experimental observations to date is the fact that the steels which show the least embrittlement generally have lower initial NDT temperatures. This observation holds for the HY-80 (NDT temperature of -190°F), the A353 (NDT of -300°F), and the A212-B steel containing 0.06% urainum (NDT of -105°F). However, the A350 steels (NDT of -40 and -80°F for two grades studied) are an exception to this observation. Also, it should be noted that the HY-80 exhibited greater embrittlement than A302-B when the irradiation temperature was 740°F. This anomaly which is thought to be the result of the combined effects of neutron embrittlement and neutron enhanced instability embrittlement is being investigated in more detail.

The Bettis study (13) comparing heats of A302-B steel also indicates the same tendency toward low sensitivity for heats having better initial notch ductility properties. Exceptions to this trend were observed there also, however.

While the variation in embrittlement response holds promise for eventually minimizing the problem, there are current derogatory aspects as well. This was pointed out in reference to the greater embrittlement of the pressure vessel steel relative to the reference heat of A302-B in one reactor radiation damage surveillance program.

(c) The benefits of elevated irradiation temperature and annealing treatments for minimizing embrittlement are positive for every steel except the HY-80. The very significant benefits of higher irradiation temperature have been demonstrated above. Annealing treatments have also been shown to be beneficial. The very significant (~40%) recovery of notch ductility observed for A212-B steel in a recent cyclic irradiation-annealing experiment (irradiation at 550°F with one intermediate annealing treatment at 800°F) indicates the potentialities of this technique.

Thus, the trend toward higher reactor operating temperatures and the possibilities presented by annealing results are very positive factors for minimizing pressure vessel embrittlement and the resultant operational problems.

Certain negative features have been recognized in each of the foregoing factors. Nevertheless, the development of greater knowledge of the influence of each factor, the conscientious exploitation of the favorable features of each, and the careful consideration of these factors in reactor design, should permit the application of carbon and alloy steels in future nuclear engineering without major concern for neutron embrittlement.

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13. ABSTRACT

Significant embrittlement of several reactor pressure vessel steels as a result of exposure to neutrons has been demonstrated by studies at the U.S. Naval Research Laboratory. Neutron embrittlement of the carbon and low-alloy steels investigated has been defined in terms of increases in the nil ductility transition (NDT) temperature. Increases in the NDT of pressure vessel steels as great as 545°F have been observed. The extent of embrittlement has been shown to depend upon the neutron exposure ($n/cm^2 > 1$ Mev), the type of steel, and the irradiation temperature.

Embrittlement relief through annealing heat treatment at temperatures above the pressure vessel operating temperature has been demonstrated. Significant embrittlement relief was observed even with multiple irradiation-annealing cycles, the extent of relief being primarily dependent upon the irradiation temperature and the subsequent annealing temperature.

Experimental data are reviewed with consideration for application to operating nuclear pressure vessel conditions. Preliminary results of power reactor surveillance and of the evaluation of one pressure vessel after nuclear service are related to experimental results. The value of extending the evaluation of

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13.

surveillance specimens and of pressure vessels after removal from nuclear service are reviewed with reference to current uncertainties as to possible nuclear environmental and stress effects.

Possibilities for using the favorable aspects of certain variable experimental factors, such as differences in embrittlement sensitivity between steels, are suggested for minimizing steel embrittlement in future reactors.