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Damaging Neutron Exposure Criteria for Evaluating the Embrittlement of Reactor Pressure Vessel Steels in Different Neutron Spectra

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ABSTRACT

Irradiation experiments to define behavior trends of reactor structural materials are frequently performed in reactor environments dissimilar to those expected during actual service. In order to accurately assess the damage produced in accelerated experimental environments so that the results can be applied to operating reactor cases, a damaging-neutron exposure criterion must be established which will account for the significantly damaging portion of the incident neutron spectra of both reactor environments. Several such exposure criteria have been evaluated through use of the results of metallurgical tests of reference steel specimens after irradiation in light and heavy water moderated reactor environments as well as in graphite moderated reactor environments.

The radiation-induced transition temperature or nil-ductility transition (NDT) temperature increases of the several steels involved are presented versus n/cm^2 determined by each of the following techniques: (a) assumption of a fission spectrum, extrapolation of activation data induced at a high Mev threshold to 1 Mev, and reporting exposure >1 Mev, and (b) calculation of spectra used to determine activation cross section for exposures above energy limits of 1, 0.5, and 0.183 Mev. The differences observed by this analysis were intercompared in relation to absolute magnitude as well as in terms of engineering significance. By applying these criteria to data relating directly to a pressurized light water power reactor, benefits to the lifetime of the reactor can be realized. The results of this study to date indicate that data relating to the properties of steels irradiated in or near the core of pressurized light water moderated reactors can be confidently intercompared for engineering applications assuming a fission spectrum and accounting for neutrons of energies >1 Mev. On the other hand, calculated spectra and average cross section adjustment to an energy limit as low as >0.183 Mev must be applied to data from highly moderated reactors.

PROBLEM STATUS

This is an interim report on one phase of the problem; work on this and other phases is continuing.

AUTHORIZATION

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DAMAGING NEUTRON EXPOSURE CRITERIA FOR EVALUATING THE EMBRITTLEMENT OF REACTOR PRESSURE VESSEL STEELS IN DIFFERENT NEUTRON SPECTRA

INTRODUCTION

The selection of different steels for application as reactor structural components is supported by the results of irradiation effects studies performed under conditions designed to represent expected reactor operations. Frequently and necessarily, these results are obtained on an accelerated basis in controlled-temperature irradiations within or near the core of a light water moderated test reactor where the expected-lifetime component neutron exposure of many years can be accumulated in a period of only weeks or months. The expected service temperature usually can be accurately controlled, and the resultant measured damage does not appear to be significantly related to the rate of accumulation (1).

For the steels being studied to develop information pertinent to reactor pressure vessels, the primary irradiation damage effect is the increase in the ductile-brittle transition temperature or the nil-ductility transition (NDT) temperature of the particular steel. This transition temperature increase (ΔNDT) is then plotted versus the neutron exposure impinging upon the steel, that is, the exposure responsible for creating the transition temperature change. By exposing these steels to various levels of neutron exposure, it has been possible to establish a trend band (2) for steel response to irradiation at specific temperatures. The trend band was outlined by NRL from about 100 data points based upon the irradiation of several types of low and medium strength steels and weldments. These irradiation experiments involved exposure of steel specimens in or near the core of light water moderated reactors. The differences in sensitivity to irradiation of these steels defined the general boundaries of the band. The neutron exposures for these irradiations were determined by the activation of iron flux monitor wires at relatively high energy thresholds by the reaction $Fe^{54}(n,p)Mn^{54}$ and extrapolation of the activation data to 1 Mev assuming a fission neutron spectral shape at the irradiation location.

The in-core test reactor neutron environment responsible for the irradiation damage defining steel behavior trends, however, is not entirely the same as that to which an in-reactor component will be exposed. Since neutron spectra are infinitely variable within and between reactors (to one degree or another), it is important to determine what portion of these spectra constitutes the significantly damaging neutron population. By providing the means for the definition and determination of this population in any given reactor location, it should be possible to obtain research results in one reactor system and confidently apply them to another reactor. Similarly, intercomparisons of test results from irradiations performed in various positions of different reactor systems could also be confidently made.

The present, widely employed neutron exposure criterion is based upon the assumption of a fission neutron spectral shape above 1 Mev at the irradiation location (Fig. 1). In practice, this is not the case, although in-core and near-core positions of light water moderated reactors appear to conform quite well to this assumption, based upon observations

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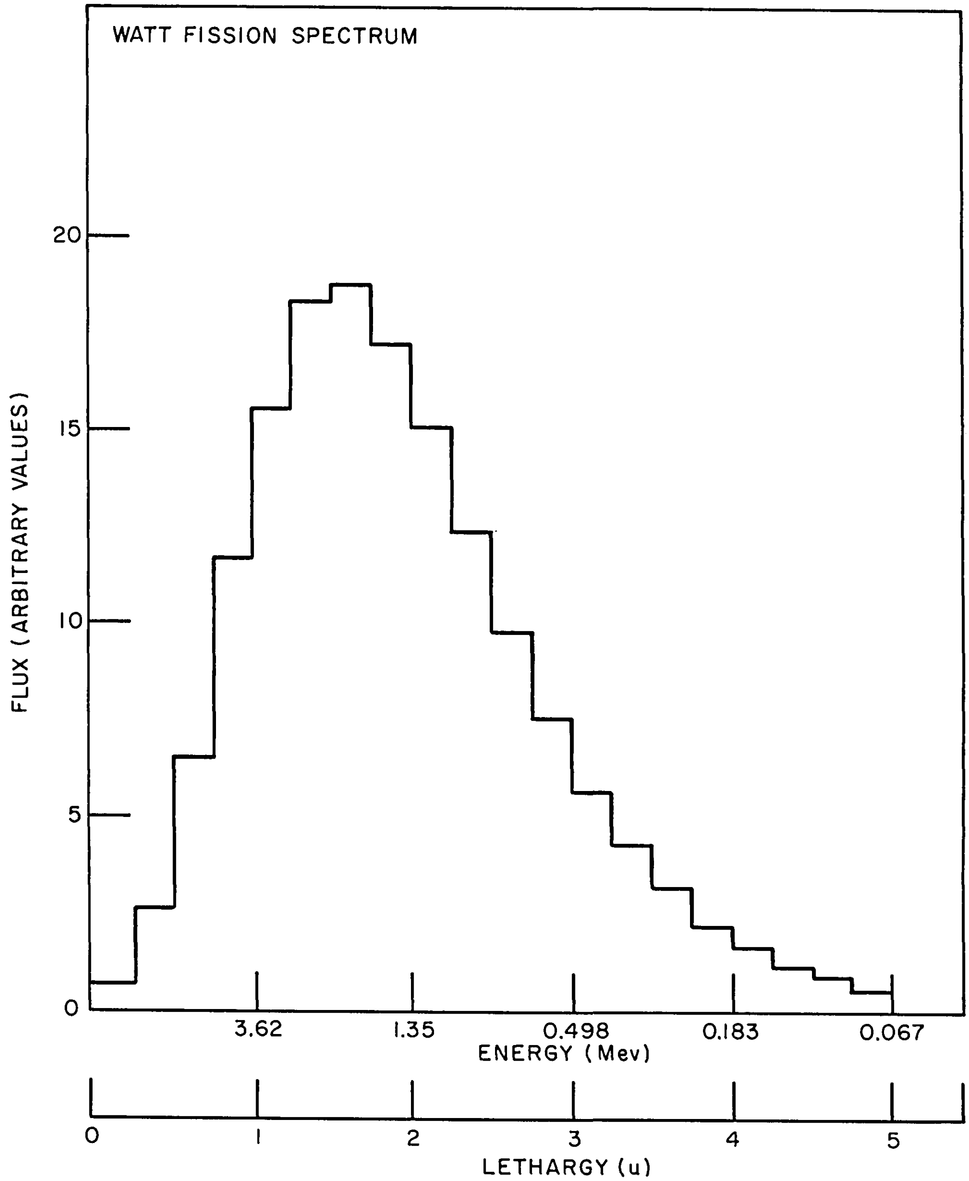

Fig. 1 - Graphical representation of the Watt fission spectrum plotted by 0.25 lethargy units in terms of $\phi(u)$

of the steel behavior trends between transition temperature increase and neutron exposure as determined by this assumption.

Neutron spectra of locations within reactors can be calculated (3-5); based upon these spectra, individual average activation cross sections for iron flux monitors (Fe^{54}(n,p)Mn^{54} reaction) can be calculated using different lower energy limits (3). The measured neutron fluxes for irradiation experiments may then be adjusted by these calculated, average cross sections and plotted versus the transition temperature increases in an attempt to reduce the variation between data points arising from exposure in different reactor environments. The more tedious nature of the approach which uses a calculated spectrum and the resulting average cross section as the technique to determine the neutron exposure, however, warrants a thorough investigation of its potential benefits before it is adopted at the expense of the simple and widely employed approach which assumes a fission spectrum at the irradiation location.

NRL has engaged in a research program to assess the benefits of the calculated exposure determination approach by comparing transition temperature (Δ NDT) changes from many irradiations in various neutron environments versus the two different exposure determination techniques. By this means, it is possible to differentiate between those reactor environments which conform well to the assumptive fission spectrum criterion, those which conform better to the calculated exposure criteria, and those which still require further investigation.

CALCULATED NEUTRON EXPOSURE CRITERIA

Neutron spectra of the NRL reactor-exposure locations have been calculated by Dahl and Yoshikawa at Battelle-Northwest Laboratories (6) using the reactor physics codes Program S by Duane (7) and Program 2DXY by Bengston et al. (8). Both of these transport-theory computer codes employ the Sn method of transport equation solution. Program S (7) has been used for symmetrical configuration calculations such as those for a reactor pressure vessel wall, since this code provides the capability for a two-dimensional analysis required for that reactor system. Program 2DXY (8) has been used for all other calculations presented in this report, as each of these reactor locations are considered asymmetrical and thus must be treated by a code which can accept input relative to dissimilar adjacent materials. Both codes were unmodified for use in the determination of spectra presented in this report.

The input for the calculations required detailed drawings and dimensions of all components from reactor core centerline to the point of interest for calculations. The composition and enrichment of fuels, as well as the composition of structural components, coolants, and moderators, were required. The codes thus required the definition of each material from the reactor core center to the point of interest.

The calculated spectra are presented in terms of flux in arbitrary values versus energy (Mev) and lethargy (u). No attempt has been made to relate experimental fluxes to the arbitrary value of the spectra.

From the neutron spectra determined as noted above, Dahl and Yoshikawa have then determined spectrum-averaged cross sections $\bar{\sigma}$ for the Fe^{54}(n,p)Mn^{54} reaction in these spectra for neutrons above lower energy limits of 1, 0.5, and 0.183 Mev (Table 1) according to the equation

$$\bar{\sigma} = \frac{\int_0^\infty \sigma_{Fe} \; \phi(E)\, dE}{\int_{E_L}^\infty \sigma(E)\; dE} \qquad (1)$$

where σ_{Fe} is the differential cross section for iron, $\phi(E)$ is the flux spectrum, and E_L is the lower energy limit: 1, 0.5, or 0.183 Mev.

The spectrum-averaged cross sections for different reactor facilities listed in Table 1 may be used directly for the determination of neutron flux from activation data or may be used to convert fluxes and exposures already determined based upon n/cm^2 > 1 Mev assuming a fission spectrum.

All of the measured neutron exposures presented in this report evolve from flux densities (referred to simply as flux) in terms of n/cm^2-sec >1 Mev assuming a fission spectrum at the irradiation location and using a fission averaged activation cross section of 68 mb. The conversion of fluxes (or integrated exposures as given in Table 1) to account for the calculated spectrum-averaged cross sections has been accomplished using

$$\frac{68\ mb}{0.692} = 98.26\ mb \qquad (2)$$

Table 1
Spectrum-Averaged Cross Sections For Mn^{54} Activation, and Thermal-to-Fast (>1 Mev Fission) Flux Ratios For Different Reactor Facilities

Reactor	Facility	ϕ 2200 m/sec / ϕ > 1 Mev (fission)	$\bar{\sigma}$ (Eq. (1) (barns)		
			E_L > 1 Mev	E_L > 0.5 Mev	E_L > 0.183 Mev
LITR	18	1.37	0.0819	0.0554	...
	28	≈1	0.0800	0.0552	...
	53	2.07	0.0736	0.0490	...
	55	1.58	0.0722	0.0487	...
	49	>2	0.0620	0.0413	...
IRL*	4-5/8†	0.973	0.1068	0.0778	0.0635
	5-5/8	0.640	0.0861	0.0586	0.0461
	6-5/8	0.537	0.0725	0.0456	0.0345
	7-5/8	0.545	0.0626	0.0367	0.0266
	8-5/8	0.850	0.0547	0.0298	0.0210
E[illegible]	W-44	22.5	0.0654	0.0388	0.0257
HWCTR	Gray Rod‡	47.0	0.1050	0.0725	0.0495
	Gray Rod§	33.5	0.1050	0.0725	0.0495
SM-1A	Pressure Vessel	...	0.1166	0.0730	0.0512
	Above Core	...	0.1615	0.1326	0.1224

* $\bar{\sigma}$ values are averages of two vertical planes through specimen positions.
†Distance from core in inches.
‡Values at location of A212-B specimens.
§Values at location of A302-B specimens.

where 0.692 is the fraction of neutrons of energies greater than 1 Mev in a fission spectrum, for use in the relation

$$\Phi(\bar{\sigma}_{E_L}) = 98.26 \frac{\Phi > 1 \text{ Mev (fission spectrum, } \bar{\sigma} \text{ 68 } mb)}{\bar{\sigma}_{E_L}} \tag{3}$$

where $\Phi(\bar{\sigma}_{E_L})$ is the integrated neutron exposure utilizing a calculated spectrum-averaged cross section above the energy limit 1, 0.5, or 0.183 Mev.

The three lower energy limits being evaluated arise from studies performed by Dahl and Yoshikawa (3) which assume that gross displacement production parallels changes in mechanical properties. This assumption suggests that whatever the defect structure may be which influences a chosen property change, accumulation of those defects will bear the same relationship to displacement production if all environmental factors other than neutron spectra are constant. Using significantly different damage models such as that of Rossin (4), which places emphasis upon high energy neutrons, and of Kinchin and Pease (5), which places more emphasis upon lower energy neutrons, they have shown that displacements in iron may be very similar in magnitude per unit of flux above a lower energy limit of about 0.4 Mev. Therefore, this exposure unit or criterion has been shown to be relatively insensitive to the damage model – the most uncertain quantity in the analysis. Further, Dahl and Yoshikawa have shown the similarity in magnitude of displacements produced by 0.4 Mev neutrons in a variety of reactor systems ranging from unmoderated fast breeder through light water to heavy water moderated reactor types. Thus, the existence of an effective lower energy limit for damage in iron which will correlate experimental results to operating reactor cases on the basis of equivalent damage production rates has been proposed. For use of this technique, it would only be necessary to employ a 0.4 Mev lower limit (0.5 Mev has been adopted for convenience) for determining the average cross section of a particular neutron spectrum for activation of Mn^{54} in that spectrum and to use this average cross section for flux and exposure determinations. Included within the Dahl-Yoshikawa analysis as well are provisions for comparisons of data based upon damage at a 1 Mev lower energy limit for comparison with the present fission spectrum >1 Mev criterion, and at a 0.183 Mev lower energy limit to assure that additional small effects from these lower energy neutrons are not unduly ignored.

EXPERIMENTAL APPROACH

In order to accurately assess the damaging potential of various reactor neutron spectra incident upon materials in experimental irradiations, it is necessary to fix all variables except the neutron spectrum. Thus in the NRL investigation, Charpy V-notch specimens of single plates of ASTM A212-B and A302-B reference steel have been irradiated at <450°F in different facilities of two light water moderated testing reactors and in a heavy water moderated testing reactor as well as in a graphite moderated research reactor. In addition, as part of the pressure vessel surveillance research conducted for the Army SM-1A reactor, Charpy V-notch specimens of A350-LF1 (Modified) plate and forging steels have been irradiated both in a light water test reactor and in the SM-1A reactor. Although not with the same materials, the concept of the investigation is being extended to higher temperatures in one other light water moderated power reactor, in an organic moderated experimental reactor as well as in an organic moderated power reactor, and in two sodium-cooled fast-breeder reactors.

IRRADIATION EXPERIMENTS

Light Water Moderated Environment

Low Intensity Test Reactor (LITR) – Charpy V-notch specimens of the ASTM reference A212-B and A302-B steel plates, sealed within capsules designed to minimize gamma heating effects, thus maintaining temperatures below 240°F, have been irradiated in five distinct core lattice positions of the Oak Ridge 3 Mw Low Intensity Test Reactor (LITR). The positions (Fig. 2) afforded experimental facilities within a partial fuel element of the

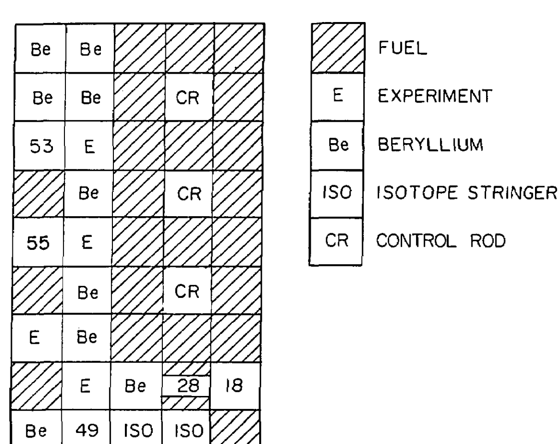

Fig. 2 - Schematic representation of the Low Intensity Test Reactor core loading during irradiation experiments. Position C-28 is a partial fuel element with provision for an experiment, while positions C-18, C-53, C-55, and C-49 are dummy fuel elements also with provision for irradiation experiments.

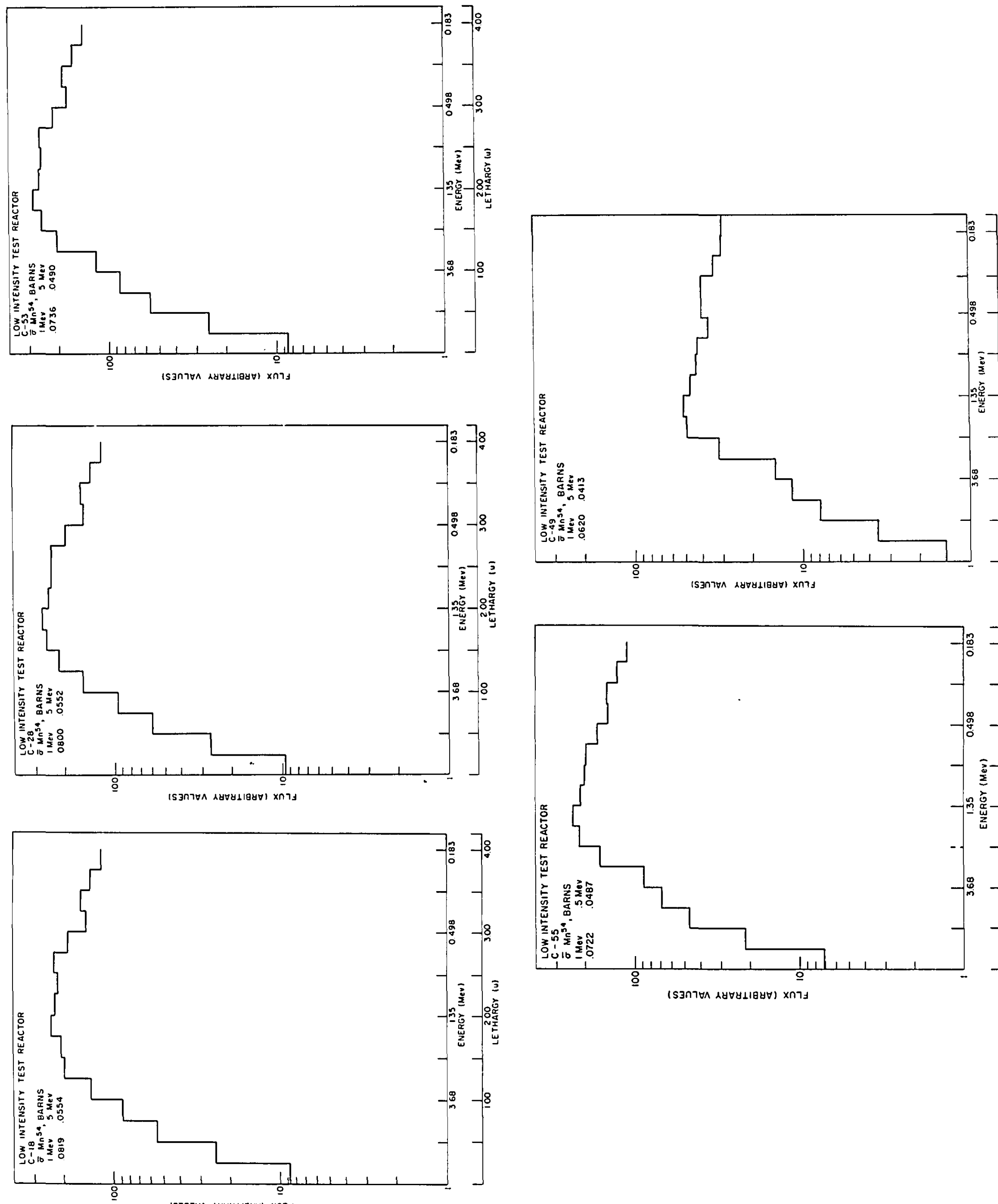

Fig. 3 - Graphical representation of the neutron spectra in the five facilities shown in Fig. 2 of the light water moderated Low Intensity Test Reactor. The iron activation cross sections for two calculated criteria are shown on each spectral plot.

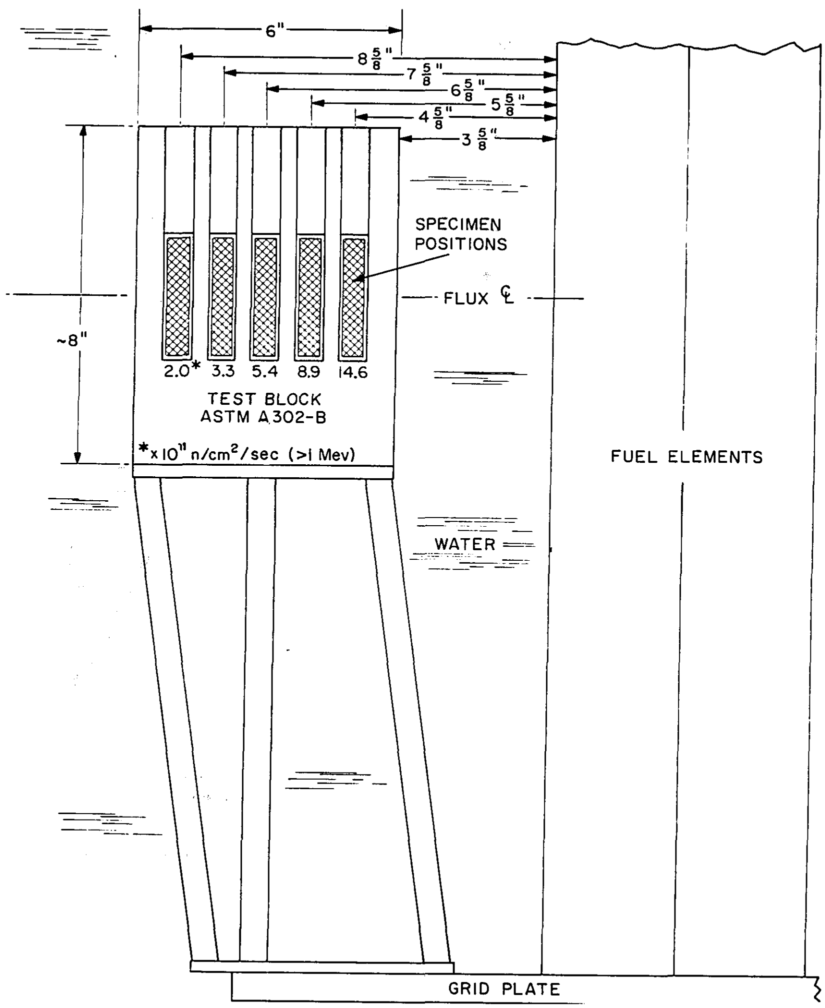

Fig. 4 - Schematic representation of the location of five specimen assemblies located in a steel block at the face of the Industrial Reactor Laboratories pool reactor

main core body (C-28), in dummy fuel elements at the boundary of the main core body (C-18), outside the main core but adjacent to fuel (C-53 and C-55), and outside the main core relatively far from any fuel (C-49). The neutron spectra for these five locations, calculated using Program 2DXY (8), are shown in Fig. 3.

Industrial Reactor Laboratories, Inc. (IRL) - Five sealed capsules each containing Charpy V-notch specimens of the ASTM reference A302-B steel were simultaneously irradiated at less than 240°F within pockets of a large steel block placed at the core face of the Industrial Reactor Laboratories (IRL) 5 Mw pool reactor (Fig. 4) (9). The spectra calculated for one vertical plane passing through the five block positions as calculated by Program 2DXY (8) are shown in Fig. 5. Spectra were actually calculated for two vertical planes passing along core lattice rows 3 and 5, extending through the block as noted on the inserts of Fig. 5. The spectra of each plane for a specimen layer location in the block (4-5/8 in., 5-5/8 in., etc., from the core face) were quite similar in shape and magnitude,

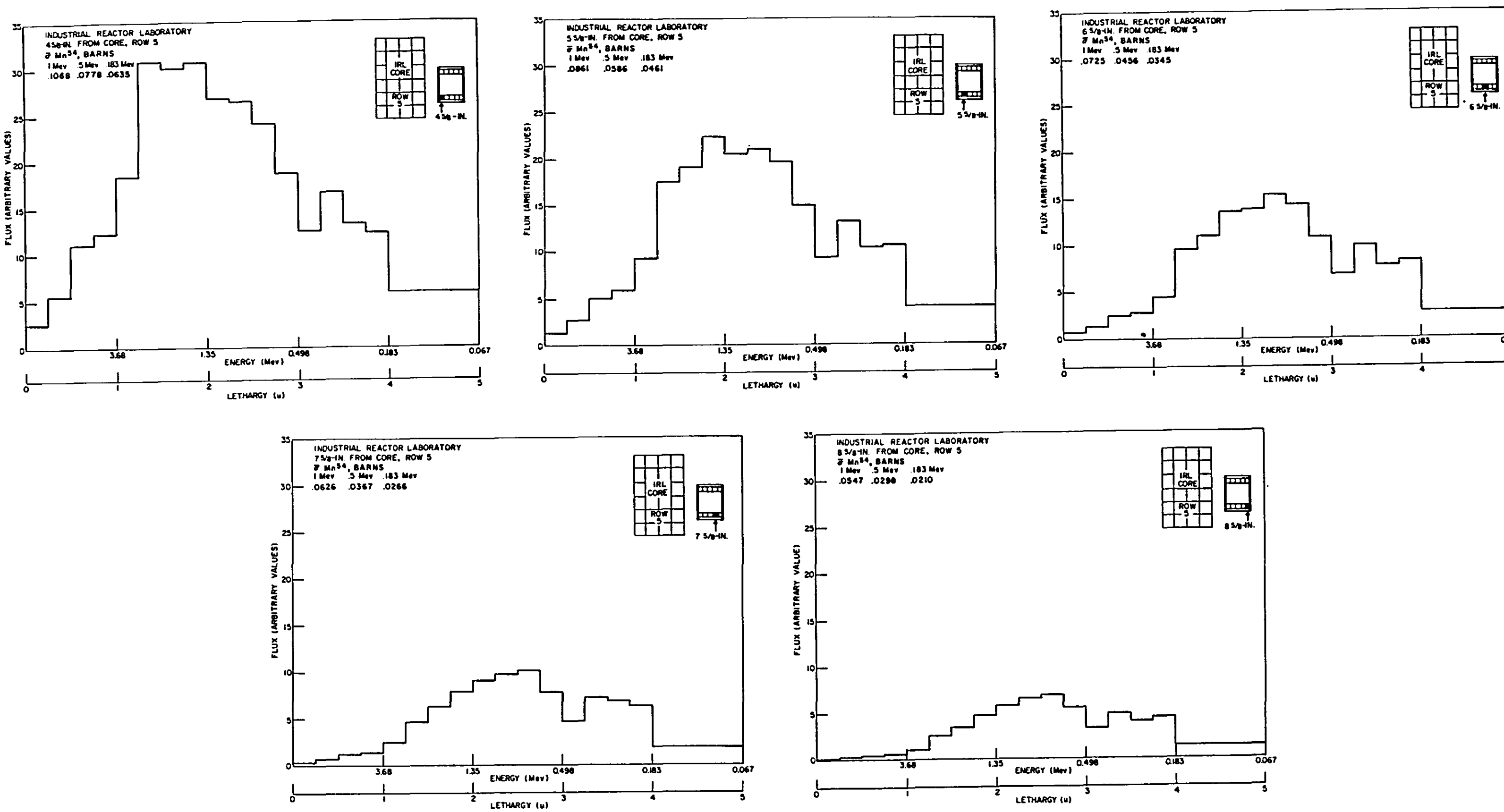

Fig. 5 - Graphical representation of the neutron spectra in the five locations shown in the schematics and in Fig. 4 of the embrittlement-in-depth test block simulating a pressure vessel wall at the face of the light water moderated Industrial Reactor Laboratories pool reactor. Two series of spectra were calculated for the test block, indicated by the two rows of positions in the schematics of each spectrum, but only one series is plotted. The iron activation cross sections given, however, are the average of the two positions at the same distance from the fuel core.

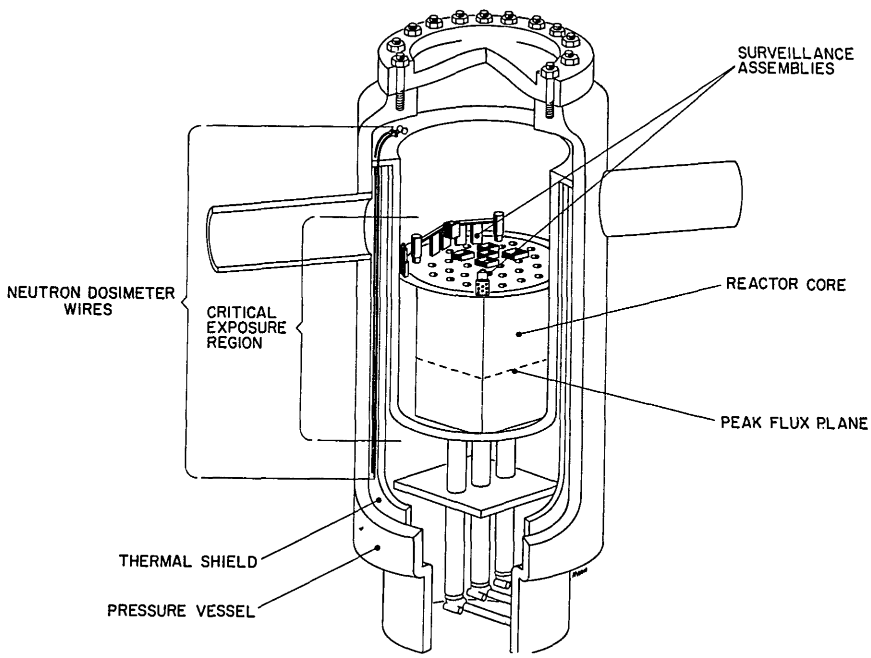

Fig. 6 - Schematic diagram of the Army SM-1A reactor showing the location of the significant parts of the reactor, the above-core surveillance assembly position, and the neutron flux monitors along the vessel wall

so that only the plane showing more moderated spectra are presented. (The average Mn^{54} cross sections noted in Fig. 5, however, are the average of the two planes.) The 6 in. thick steel block simulated a section of a pressure vessel wall, with the experimental assembly pockets positioned through the thickness of the block (from 4-5/8 in. to 8-5/8 in. from the core face) for simplicity in placing the specimen-containing assemblies for irradiation to measure embrittlement through the block thickness.

Army SM-1A Reactor - Charpy V-notch specimens of the A350-LF1 (Modified), SM-1A pressure vessel steel, were irradiated at temperatures between 445° and 475°F within four sealed capsule assemblies suspended along an arc at the upper edge of the SM-1A reactor core (Fig. 6). The spectrum for this location (Fig. 7) was calculated using Program S (7). (Accelerated irradiations of this steel have also been performed at 430°F in the LITR.) Flux monitors (neutron dosimeter wires) were located along the inner edge of the vessel wall (10) (Fig. 6); the spectrum of this position (Fig. 8) was calculated using Program S (7).

Graphite Moderated Environment

Brookhaven Graphite Reactor (BGR) - Two sealed capsules each containing ASTM reference A212-B and A302-B steel Charpy V-notch specimens were irradiated at less than 280°F in W-44, a horizontal channel normal to and centered above a fuel element channel in the Brookhaven Graphite Reactor (BGR). The spectrum of this irradiation location is presented in Fig. 9 as calculated by Program 2DXY (8). One experiment was exposed for about ten months, while the other was exposed for about thirteen months.

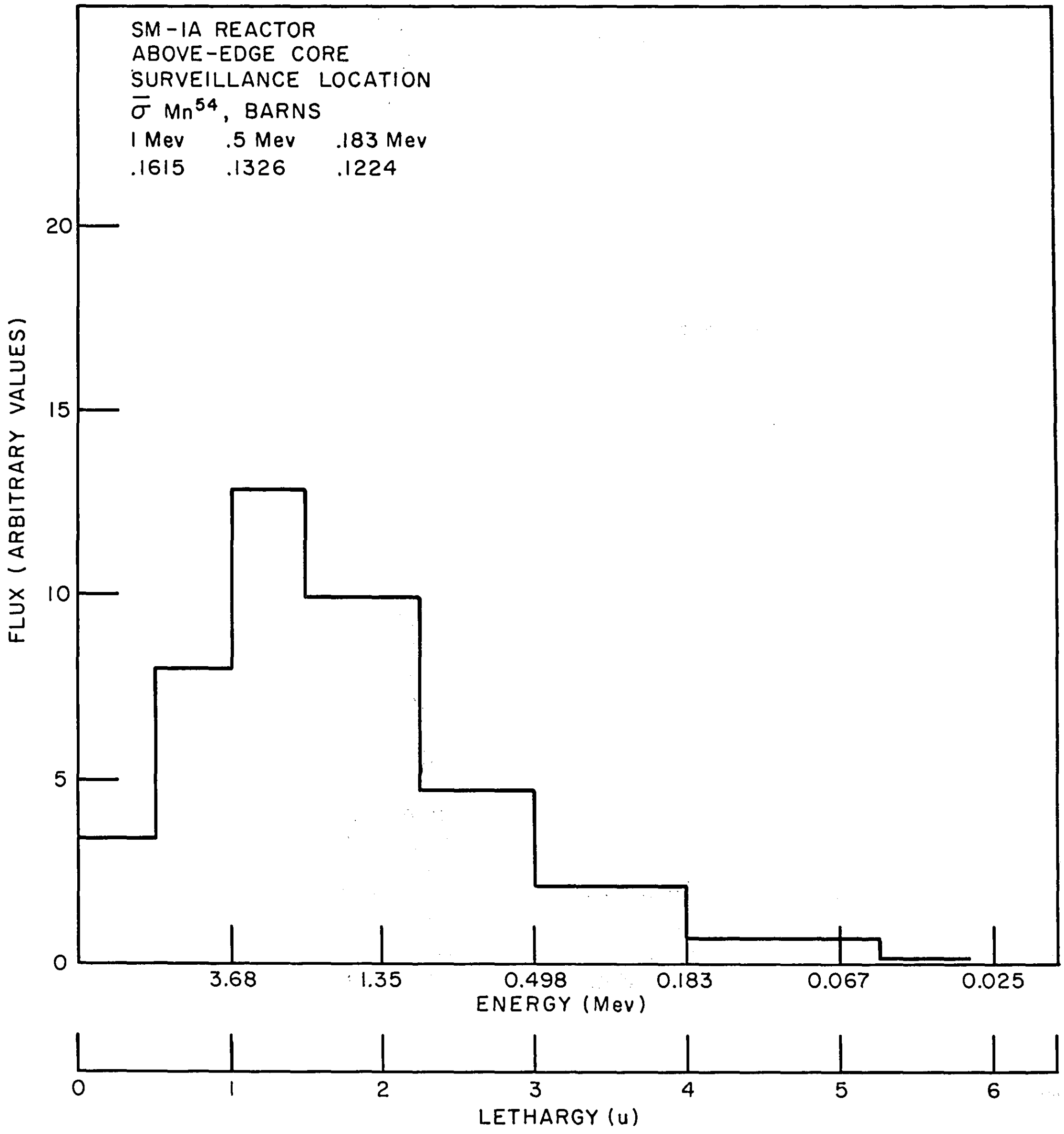

Fig. 7 - Graphical representation of the neutron spectrum at the V-notches of surveillance Charpy specimens suspended at the edge of the upper side of the light water moderated SM-1A reactor core as shown in Fig. 6

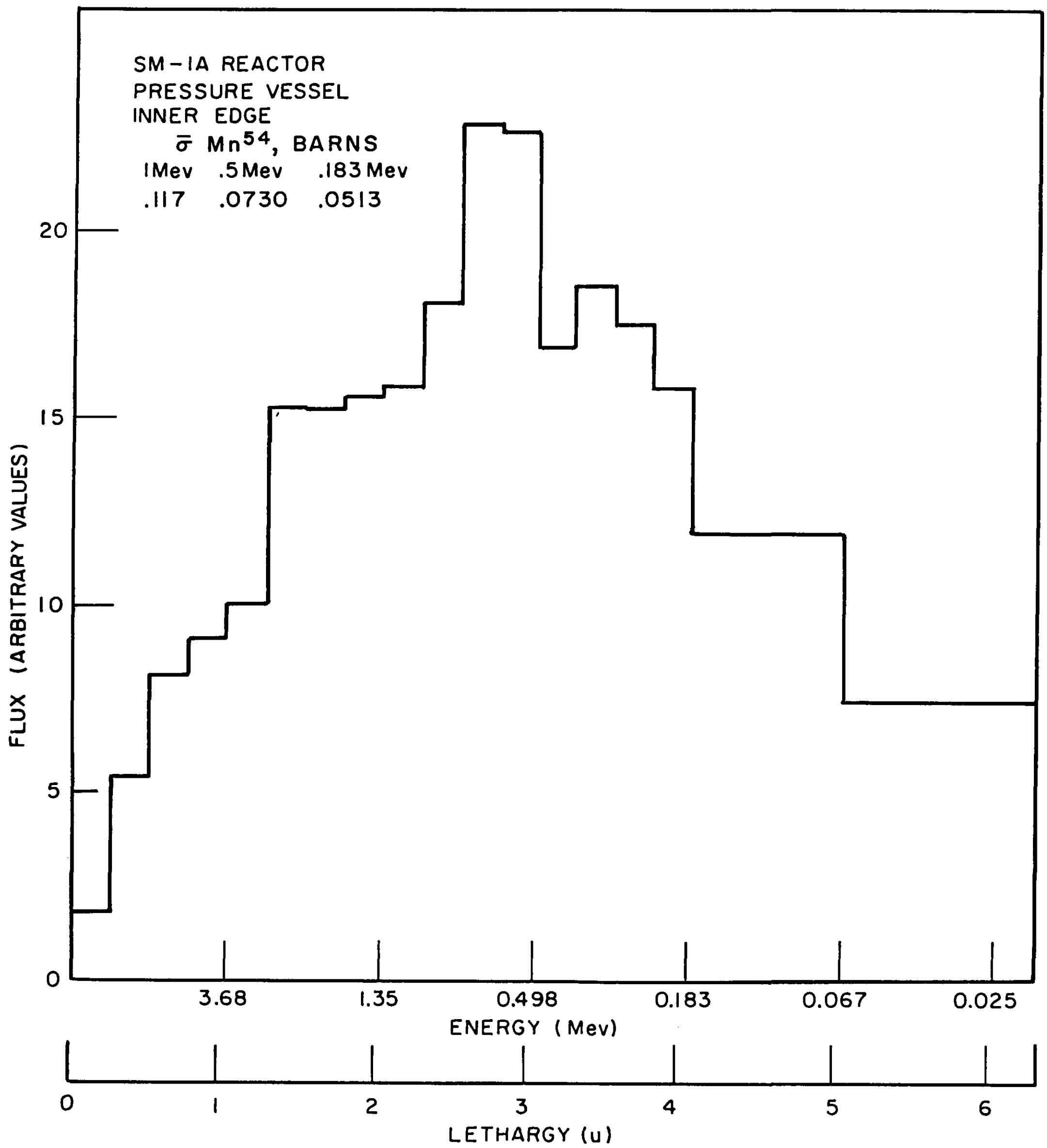

Fig. 8 - Graphical representation of the neutron spectrum at the pressure vessel wall of the light water moderated SM-1A reactor

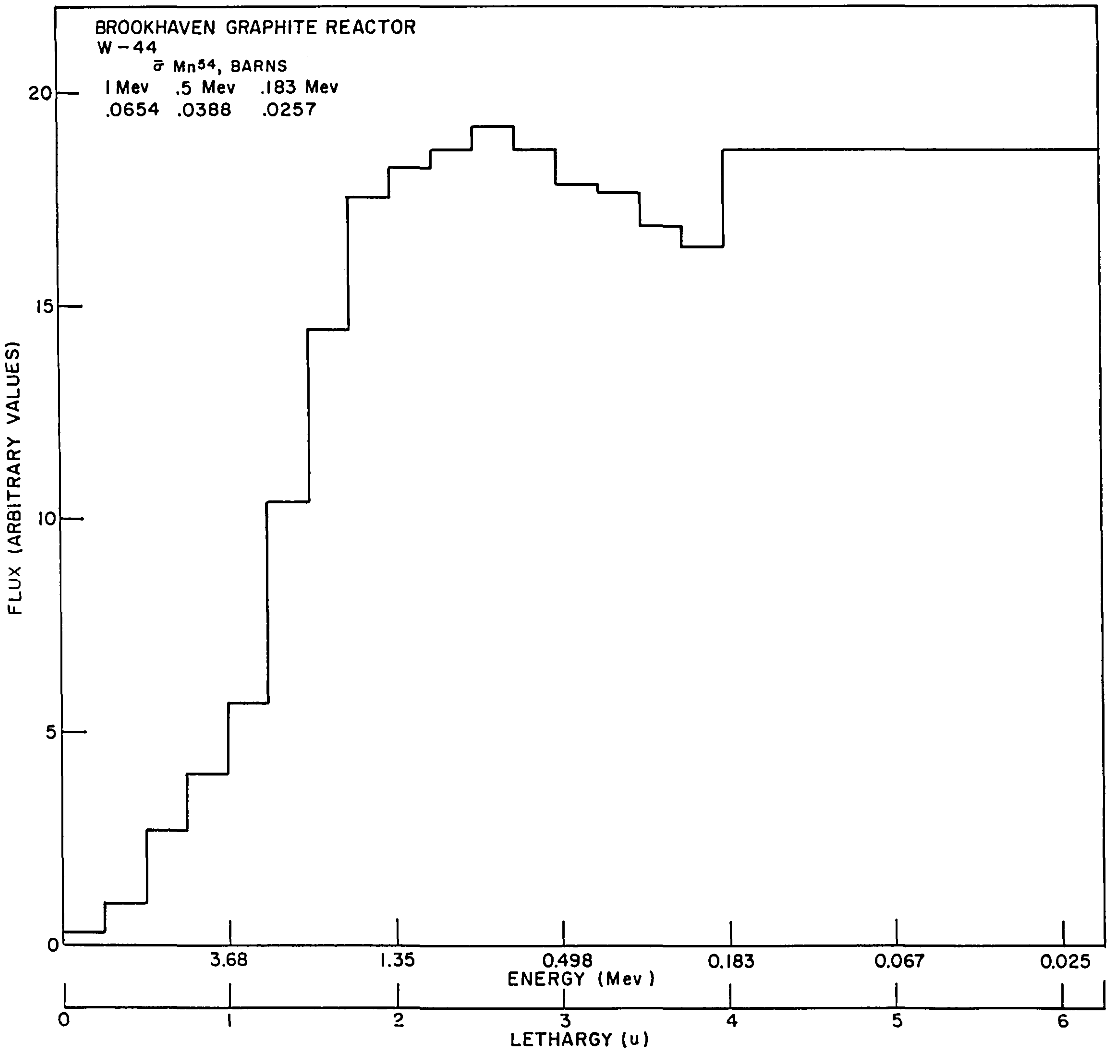

Fig. 9 - Graphical representation of the neutron spectrum above a fuel channel in the W-44 position of the Brookhaven Graphite Reactor

Heavy Water Moderated Environment

Heavy Water Components Test Reactor (HWCTR) – A capsule assembly consisting of individual sections containing A212-B and A302-B Charpy V-notch specimens aligned in a string was irradiated at less than 464°F in the central control rod cluster, gray rod position, of the 45 Mw Heavy Water Components Test Reactor (HWCTR) (Fig. 10). The 40 in. long assembly was centered 18 inches above the peak flux plane of the 120 in. high fuel core, at which point the spectrum of Fig. 11 was determined by Program 2DXY (8).

POSTIRRADIATION EVALUATION

Following each experimental irradiation the specimen capsules were returned to NRL for disassembly, removal, and testing of specimens, for the inspection of temperature monitors to assure that the proper exposure temperature limits had not been

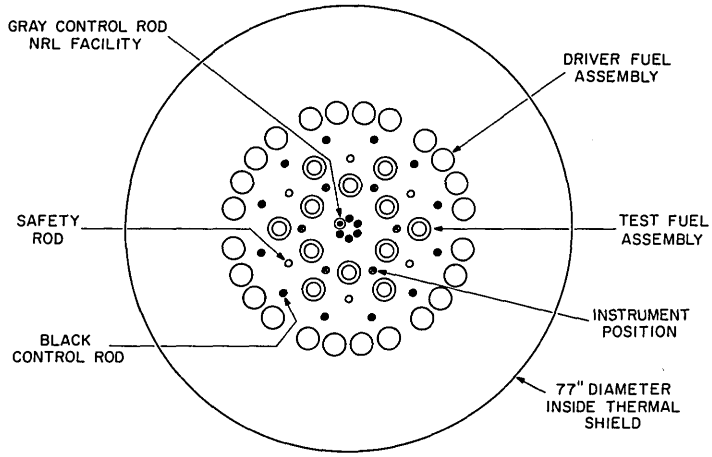

Fig. 10 - Schematic representation of the Heavy Water Components Test Reactor core arrangement showing the location of the gray rod position employed by NRL for an experimental irradiation

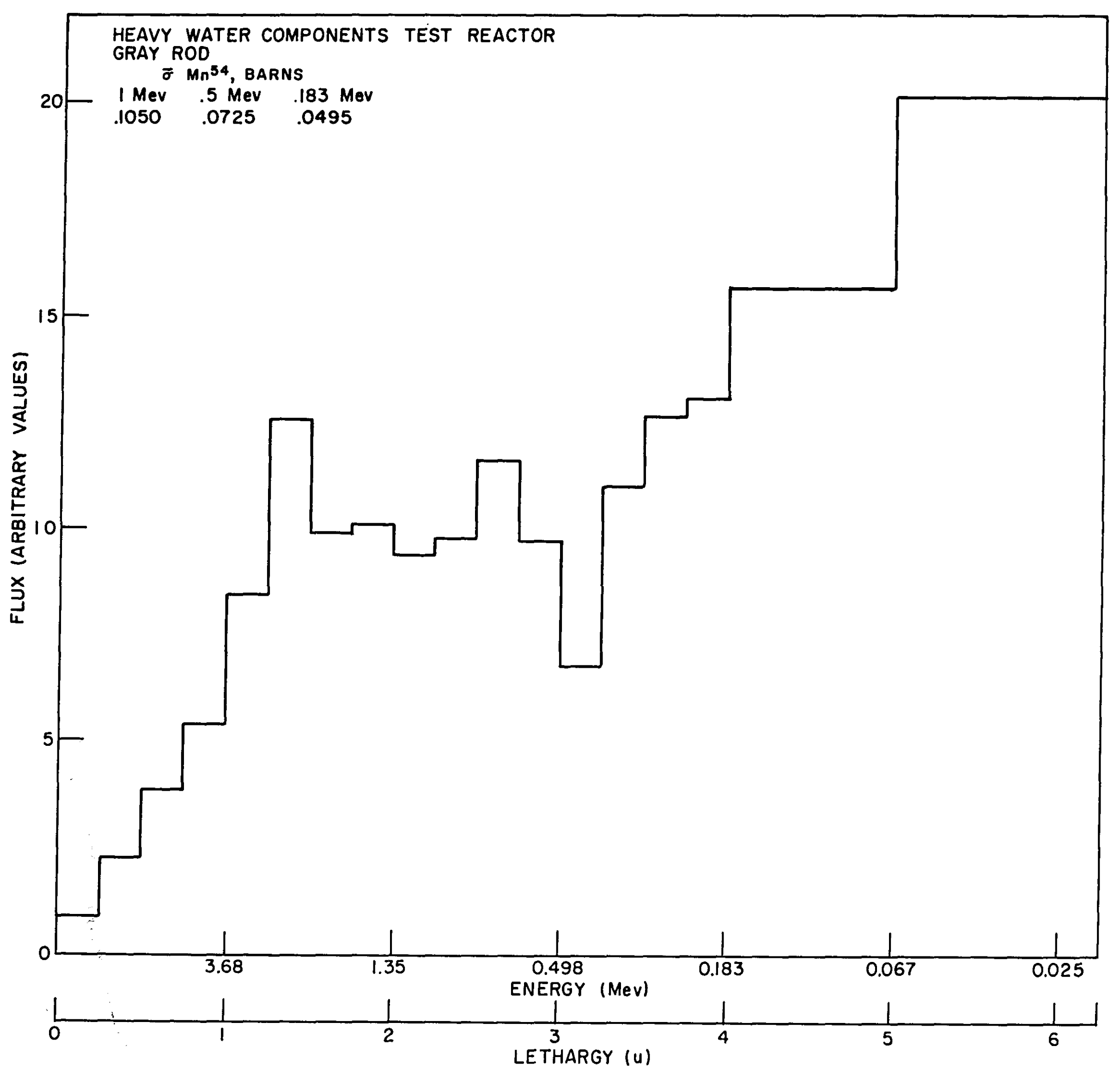

Fig. 11 - Graphical representation of the neutron spectrum 18 in. above the peak flux plane of the central control rod cluster gray rod position of the Heavy Water Components Test Reactor

Table 2
Summary of Data For The Exposure of 4-in. A212-B Steel

Reactor	Facility	Irradiation Temperature (°F)	Transition Temperature Increase (°F)	Φ (t) (n/cm^2 > 1 Mev) Fission Spectrum $\bar{\sigma}$ 68 mb Mn^{54}
LITR	18	260	210	0.66×10^{19}
LITR	18	400	180	0.66×10^{19}
LITR	18	450	200	0.66×10^{19}
LITR	18	200	185	0.78×10^{19}
LITR	18	200	215	1.0×10^{19}
LITR	18	275	290	2.1×10^{19}
LITR	18	200	295	2.5×10^{19}
LITR	18	430	305	2.6×10^{19}
LITR	28	>240	245	1.29×10^{19}
LITR	28	200	275	2.70×10^{19}
LITR	53	>240	245	1.10×10^{19}
LITR	53	>240	245	1.22×10^{19}
LITR	53	>240	280	1.30×10^{19}
LITR	55	>240	180	0.75×10^{19}
LITR	55	>240	205	0.80×10^{19}
LITR	55	200	245	0.95×10^{19}
LITR	49	>240	210	0.67×10^{19}
LITR	49	>240	200	0.70×10^{19}
LITR	49	>240	215	0.78×10^{19}
BGR	W-44	>280	255	0.55×10^{19}
BGR	W-44	>280	260	0.75×10^{19}
HWCTR	Gray Rod	>464	175	0.35×10^{19}

exceeded, and for the recovery and analysis of neutron flux monitors. Transition temperature increases of the irradiated specimens were determined using the preirradiation properties of Charpy V-notch specimens of the unirradiated parent plates. Temperature monitors for each capsule consisted of low melting point eutectic alloys sealed in quartz tubes encompassing a range of temperatures consistent with those expected for that capsule exposure. Neutron fluxes from iron, nickel, titanium, and cobalt monitor wires included in the experimental assemblies were determined by assuming a fission spectrum at each irradiation location. The calculated neutron exposures have been determined from the fission spectrum >1 Mev fluxes (exposures) using Eqs. (2) and (3).

Pertinent data from exposures of A212-B steel in all environments are presented in Table 2, while data from exposures of A302-B and A350-LF1 (Modified) steels are presented respectively in Tables 3 and 4.

DISCUSSION

There are two purposes in using calculated neutron spectra and activation cross sections. First, by accounting for the neutron population above a lower energy limit which has been shown to produce relatively equal damage rates in diverse reactor systems, data from such systems should emerge in a uniform pattern. Second, by considering the actual neutron spectra of irradiation facilities, it should be possible to better account for the damage inflicted by the actual neutron populations of these facilities and thus provide a means for more confident interrelation of data between diverse reactor exposures.

Table 3
Summary of Data for the Exposure of 6-in. A302-B Steel

Reactor	Facility	Irradiation Temperature (°F)	Transition Temperature Increase (°F)	$\Phi(t)$ (n/cm^2 > 1 Mev) Fission Spectrum $\bar{\sigma}$ 68 mb Mn^{54}
LITR	18	260	170	0.5×10^{19}
LITR	18	400	130	0.5×10^{19}
LITR	18	450	140	0.5×10^{19}
LITR	18	200	200	0.6×10^{19}
LITR	18	<240	255	1.8×10^{19}
LITR	28	<240	220	1.24×10^{19}
LITR	53	<240	215	1.20×10^{19}
LITR	53	<240	295	1.94×10^{19}
LITR	53	<240	315	2.00×10^{19}
LITR	53	<240	330	3.50×10^{19}
LITR	55	<200	230	0.85×10^{19}
LITR	49	<240	205	0.75×10^{19}
IRL	4-5/8	<240	105	0.26×10^{19}
IRL	5-5/8	<240	80	0.16×10^{19}
IRL	6-5/8	<240	50	0.10×10^{19}
IRL	7-5/8	<240	50	0.06×10^{19}
IRL	8-5/8	<240	35	0.04×10^{19}
BGR	W-44	<280	205	0.55×10^{19}
BGR	W-44	<280	240	0.75×10^{19}
HWCTR	Gray Rod	<464	190	0.73×10^{19}

Table 4
Summary of Data for the 430°F Exposures of Army SM-1A Pressure Vessel Steel – A350-LF1 (Modified) Plate and Forging

Reactor	Facility	Steel	Transition Temperature Increase (°F)	$\Phi(t)$ (n/cm^2 < 1 Mev) Fission Spectrum $\bar{\sigma}$ 68 mb Mn^{54}
LITR	18	Plate	330	2.0×10^{19}
LITR	18	Forging	340	2.0×10^{19}
LITR	18	Forging	380	2.8×10^{19}
LITR	55	Flate	415	2.8×10^{19}
LITR	18	Plate	395	3.1×10^{19}
LITR	55	Plate	440	3.1×10^{19}
SM-1A	Above Core 445-475°F	Plate	80	0.26×10^{19}

The ability of the calculated spectra-and-cross-section criteria to satisfy the two purposes will now be evaluated by reference to the A212-B and A302-B steel embrittlement data for the first purpose and by reference to the A350-LF1 (Modified) steel embrittlement data for the second.

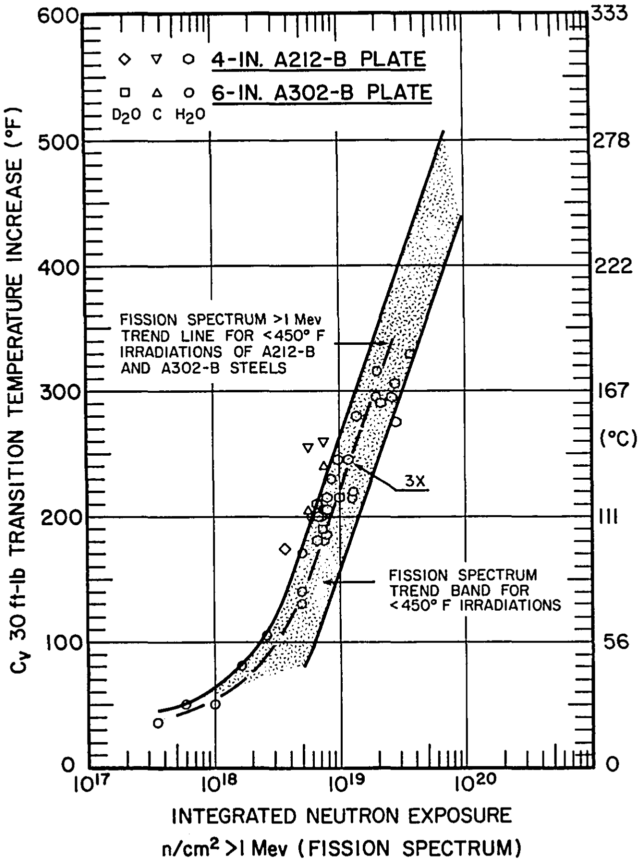

Fig. 12 - Trend line for <450 °F irradiations of A212-B and A302-B steels plotted versus neutron exposure >1 Mev assuming a fission spectrum at the irradiation locations. Data are from exposures in light water, heavy water, and graphite moderated reactors.

A212-B and A302-B Exposures

Transition temperature increases for A212-B (Table 2) and A302-B (Table 3) steels irradiated in the light and heavy water as well as graphite moderated reactor environments have been plotted versus integrated neutron exposure >1 Mev assuming a fission spectrum in Fig. 12, versus neutron exposure from calculated spectra-and-cross-sections >1 Mev in Fig. 13, and versus neutron exposure and calculated >0.5 Mev in Fig. 14. In each figure, the trend band for irradiations at <450°F as plotted by the fission spectrum criterion has been included only for reference.

Progressing from the assumed fission spectrum plot (Fig. 12) through to the calculated >0.5 Mev plot (Fig. 14), a shift to the right is noted in the average trend line for A212-B and A302-B steel response to irradiation.

When one considers the fairly large differences in spectra and calculated average cross sections between the diverse reactor spectra, the data point shift effected by the calculated >0.5 Mev criterion is not particularly large, and the shift effected by the calculated >1 Mev criterion is even less. Note that relative to the light water moderated data points, which represent the majority of the data, neither the graphite nor the heavy

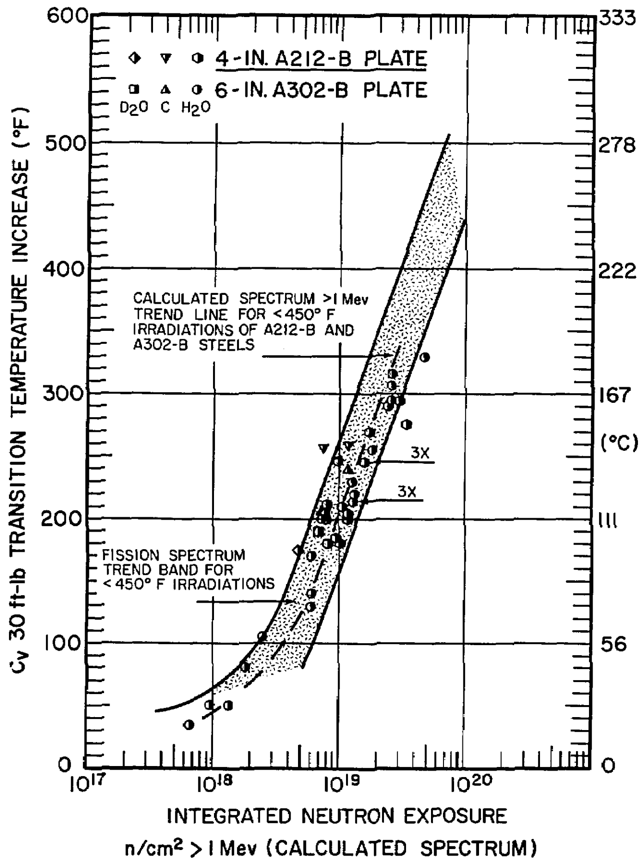

Fig. 13 - Trend line for <450°F irradiations of A212-B and A302-B steels plotted versus neutron exposure >1 Mev using calculated spectra-and-cross sections for the irradiation locations. Data are from exposure in light water, heavy water, and graphite moderated reactors.

water moderated reactor data points are shifted into significantly better overall agreement. This is quite interesting since on a semilog plot it might be expected that much better normalization or narrowing of the scatter would occur, especially for those data points from the more heavily moderated exposures. Thus the non-light-water-moderated exposures do not appear to be satisfactorily treated by the fission spectrum >1 Mev criterion. Further, if the calculated spectra-and-cross-section approach were carried to lower energy limit >0.183 Mev, virtually the same spread in data points would still be observed. It may be fair, then, to say that at least for in-core or near-core light water moderated exposures, rather good interagreement of data can be effected simply by using the fission spectrum assumption for a neutron exposure criterion.

As noted above, the data point scatter in moving from the fission spectrum criterion to the calculated >0.5 Mev criterion is not noticeably improved. While this may not be clearly seen in the semilog data plots, it does become apparent when the data of Figs. 12 and 14 are replotted on a linear scale. Figures 15 and 16 show these data, for A212-B and A302-B steel respectively, versus the fission spectrum >1 Mev and calculated >0.5 Mev criteria. Again, the scatter of data points about the two trend lines in both figures does not appear to be less for the calculated >0.5 Mev criterion. Although from an intuitive standpoint, this criterion should provide better overall normalization of data between reactors, such proof has not been established by the A212-B and A302-B steel data.

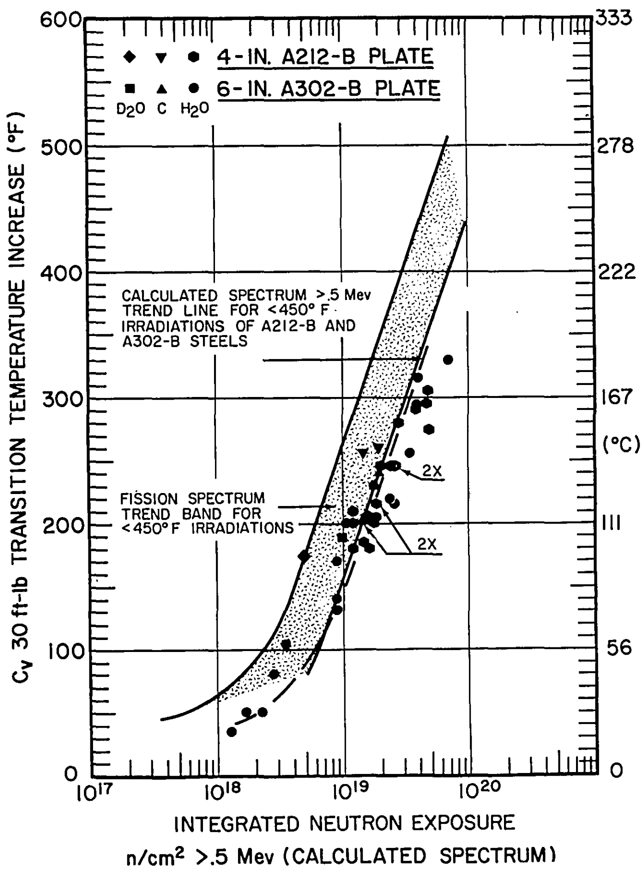

Fig. 14 - Trend line for <450°F irradiations of A212-B and A302-B steels plotted versus neutron exposures >0.5 Mev using calculated spectra-and-cross sections for the irradiation locations. Data are from exposures in light water, heavy water, and graphite moderated reactors.

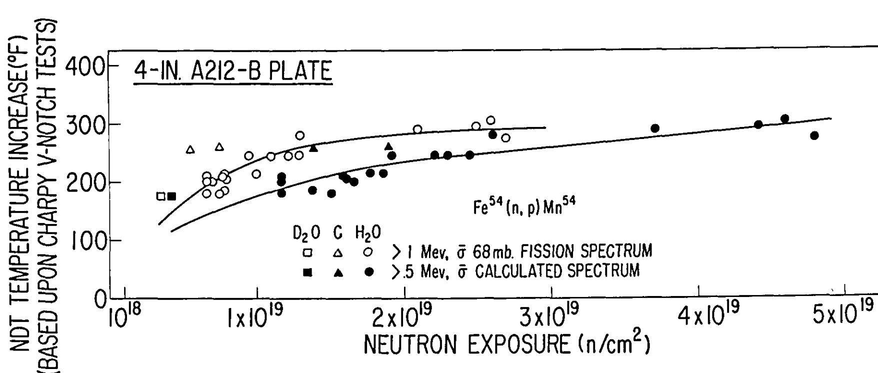

Fig. 15 - NDT temperature increase for A212-B steel versus integrated neutron exposure >1 Mev determined by assuming a fission spectrum at the irradiation location (open points) and by calculated spectra-and-cross sections >0.5 Mev (closed points).

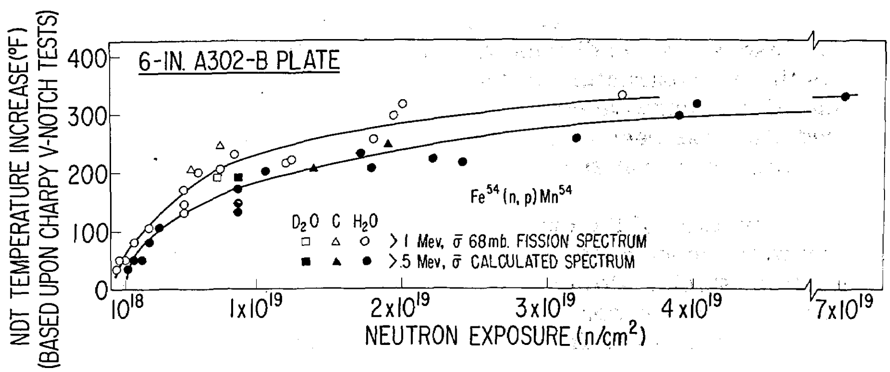

Fig. 16 - NDT temperature increase for A302-B steel versus integrated neutron exposure >1 Mev determined by assuming a fission spectrum at the irradiation location (open points) and by calculated spectra-and-cross-sections >0.5 Mev (closed points)

Closer inspection of the heavy water and graphite moderated reactor exposure data points reveals another aspect which may be of much more significance than neutron spectral effects. In Fig. 12, the A212-B steel data points from these two environments are significantly to the left of the average trend line for radiation response behavior of that steel. With respect to the graphite reactor data, by following down the respective neutron exposure ordinates for both of the A212-B data points, one finds the A302-B data points from the same irradiations to be very close to the left boundary of the trend band. It is important to note that specimens of both of these steels were simultaneously irradiated, in two separate experiments, so that both steels received the same neutron exposure within each experiment. As shown above, use of the calculated >1 Mev and >0.5 Mev criteria does not significantly improve the disparity between the graphite and light water moderated data points.

With respect to the heavy water reactor data, the A212-B data point lies well to the left of the average trend line in Fig. 12, while the A302-B data point lies very close to the trend line. The difference in exposure of these two data points results from the relatively large distance between specimen sections along the length of the experimental assembly in the reactor. Observing the relative locations of the two data points when plotted by the calculated criteria in Figs. 13 and 14 versus those plotted in Fig. 12, the effect of the very large spectral difference in the heavy water moderated reactor can be discerned. That is, the data points for the calculated >1 Mev criterion shift to the left indicating less calculated >1 Mev flux than fission spectrum >1 Mev flux. The data point shifts by the calculated >0.5 Mev criterion are then moved back just to the right of the original fission spectrum >1 Mev plot locations. The net effect is that the data points from the heavy water moderated reactor are significantly unimproved by the two calculated criteria, which was also the case for the graphite moderated reactor data. What emerges then is evidence that a factor related to the variable sensitivity of steels to radiation embrittlement apparently is of greater significance in radiation effects than adjustments for variations in the pertinent neutron spectra.

The various degrees of sensitivity of steels or even different heats of the same steel to irradiation has previously been observed and reported (11,12). These studies have shown that for a given neutron exposure in a given neutron environment, a wide range of transition temperature increases can be exhibited by these different steels and heats of steels. A recent study (13) has shown that a steel sensitive to radiation embrittlement can be reheat-treated so as to make it relatively insensitive, and vice versa. In this particular case (13), the microstructure producing high sensitivity in the two steels investigated was the same for both steels. Likewise, the microstructures for both steels in their respective insensitive conditions were also identified and found to be the same, but for both steels the microstructure of the insensitive condition was noted to be quite different from that of the sensitive conditions. Thus, a metallurgical factor influencing sensitivity to irradiation has been established and, as indicated in this report, appears to significantly outweigh spectral sensitivity considerations.

The significant deviation of the data points for the A212-B steel after irradiation in graphite and heavy water moderated reactors has thus far been attributed to possible material differences in steels. However, irradiation temperature is also a factor which must be considered as being responsible for these unusual effects. The majority of data points in Figs. 12 through 16 were from irradiations conducted at temperatures below 250°F. The heavy water moderated exposure data points, however, were from an irradiation conducted at temperatures below 464°F as determined by low melting point alloys. This temperature, although considered to be appropriate for inclusion into a radiation effects trend for irradiations below 450°F, is significantly higher than that for the rest of the data. The temperature consideration, however, tends to fail when one recognizes that both the A212-B and A302-B steels were irradiated at this same temperature, and that one steel conforms to the general trend while the other does not. A similar disparity was noted in the response of the two steels to the graphite reactor exposure, where the irradiation temperature was less than 280°F. This temperature range is well within the <450°F criterion and close to the temperature for the light water irradiations, where no such disparity between A212-B and A302-B steel data points is evident. Thus, while temperature of irradiation appears to be perhaps not a greatly significant factor in the data of this report, it still remains a factor for consideration.

In connection with the relative differences between these two steels arising from irradiations in graphite and heavy water moderated reactors, it should be noted that the data points generally lie to the left of the average trend lines for the steels described by the three exposure criteria. Since both reactor environments are highly thermalized, it appears that the preponderance of low energy neutrons may have some additional effect other than the usually recognized displacement-type damage effect.

Wechsler (14) has shown that for the same flux of fast neutrons as thermal neutrons in a fission spectrum, fission neutron collisions produce a factor of 150 more displacements than do thermal neutrons by (n,γ) recoil. Further, if the effect of anisotropic neutron scattering is included, the above factor is reduced to about 85. He concludes, then, that if the fast neutron spectrum is moderated, the factor will be reduced further, so it is apparent that reactor environments might well exist for which it is important to consider the contribution made by (n,γ) recoil displacements in producing changes in properties upon irradiation. It can be seen from the thermal-to-fast neutron flux ratios in Table 1 that the in-core or near-core light water moderated reactor facilities should not be significantly affected by (n,γ) recoil reactions, since the two neutron populations are not vastly different. Conversely, the very high thermal-to-fast neutron flux ratios for the graphite and heavy water moderated reactor facilities indicate a very real potential for some effect by (n,γ) recoil reactions upon the property changes in the steels reported in this study. This has been shown to be highly probable by Sheely (15) in a recent independent analysis upon the body of data presented in this report.

Sheely has shown, using the thesis presented by Wechsler (14), that (n,γ) recoil events would add a factor of about 1 to 3 percent to the light water moderated exposures but a factor of about 10 to 35 percent to data from the non-light-water-moderated environments used in this study.

The foregoing discussion with reference to the first purpose for using calculated spectra-and-cross-sections has indicated that a significantly better overall normalization of data from diverse reactor exposures of different steels has not been effectively realized.

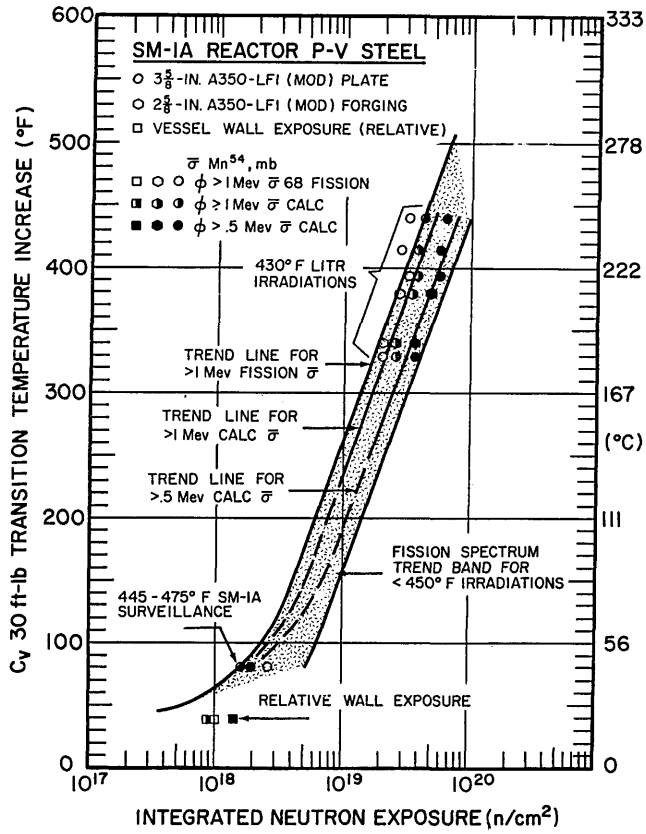

Fig. 17 - Trend line for LITR and SM-1A irradiations of A350-LF1 (Modified) steel plotted versus neutron exposures (n/cm^2) >1 Mev, assuming a fission spectrum, and by using calculated spectra-and-cross-sections for determining neutron exposures (n/cm^2) above lower energy limits 1 Mev and 0.5 Mev. The square data points show the relative vessel wall fluxes per unit of time as plotted by the three exposure criteria and do not relate to any particular time in the life of the reactor.

Better knowledge of the contribution of three factors, namely, the sensitivity of steels to radiation embrittlement, temperature effects, and the very-low-energy-neutron (n,γ) recoil damage, may ultimately provide some additional clarification which will hopefully improve the overall normalization.

A350-LF1 (Modified) Steel Exposures

The second purpose for using calculated neutron spectra criteria is to better account for the actual neutron environment to which a steel is exposed and thereby provide a basis for more confident interrelation of data between diverse reactor exposures. Fulfillment of this purpose is satisfied by consideration of the transition temperature increase data for the Army SM-1A reactor pressure vessel steel, A350-LF1 (Modified). This data is plotted in Fig. 17 versus the three exposure criteria. The trend band in Fig. 17, plotted using the assumed fission spectrum criterion, is included only for reference. The higher neutron exposure points were obtained from irradiations at 430°F in positions C-18 and C-55 of the LITR, while the 80°F transition temperature increase data point was based upon specimens from an above-core surveillance exposure position in the SM-1A reactor.

When all of the SM-1A reactor data is plotted in terms of either of the calculated criteria, the significant differences in neutron spectra of the various irradiation locations can be easily observed, and a more encompassing treatment of the data can be made as follows.

The open data points in Fig. 17 represent measured transition temperature increases resulting from controlled temperature, 430°F LITR and 445-475°F SM-1A irradiations as plotted by the assumed fission spectrum >1 Mev criterion. The LITR data points at relatively high transition temperature increases fall along the left side of the assumed fission spectrum trend band; the SM-1A surveillance data point falls about midway within the band. The embrittlement limit as related to the life of the reactor vessel is concerned with higher increases. Thus, the trend line which has been constructed for irradiation embrittlement response of the Army A350-LF1 (Modified) steel in terms of n/cm^2 >1 Mev is defined by the LITR data points and corresponds to the left side of the fission spectrum trend band.

The next step in the analysis is in recognition of the neutron spectral differences between the LITR and SM-1A reactor irradiation locations and entails the replotting of transition temperature increase data points in terms of n/cm^2 >1 and >0.5 Mev, using calculated spectrum-activiation cross sections for the respective reactor positions. The half closed and fully closed data points in Fig. 17 define the trend for these irradiations as determined respectively by the calculated > 1 Mev and > 0.5 Mev exposure criteria. It can be seen that the higher exposure data points in the LITR spectra shift to the right, indicating more calculated flux in the LITR spectra than in an assumed fission spectrum. This is graphically illustrated in Fig. 18. Conversely, the spectrum at the SM-1A surveillance location (80°F transition temperature increase) indicates less calculated spectrum flux than fission spectrum >1 Mev flux; thus the data points are shifted to the left. The trend line for calculated > 0.5 Mev behavior of the A350-LF1 (Modified) steel, described by the closed data points, can be noted to indicate that almost 90 percent more calculated >0.5 Mev flux is required in the LITR irradiations to effect the noted transition temperature increases than would be required on the basis of assumed fission spectrum >1 Mev flux. This becomes reasonable by noting that the LITR C-55 and C-18 spectra (Fig. 3) are more intense in the region below 1 Mev than is the Watt fission spectrum of Fig. 1. Further, as shown in Fig. 18, the LITR spectra have almost twice the neutron population above 0.5 Mev than does the fission spectrum above 1 Mev, while the SM-1A wall spectrum had about one-and-one-third the neutron population above 0.5 Mev than does the fission spectrum above 1 Mev.

The irradiation behavior of A350-LF1 (Modified) steel, described by the trend lines in Fig. 17, may now be related to embrittlement at the peak flux plane of the SM-1A pressure vessel wall for the three exposure criteria using Fig. 18. If one assumes the instantaneous neutron flux in the LITR to be equivalent to that in the SM-1A and uses the fission spectrum >1 Mev exposure criterion, the exposure time required to produce a transition temperature increase will be the same, and the trend line at the left side of the band in Fig. 17 will govern the lifetime embrittlement behavior of the steel.

For the assessment of radiation behavior in terms of the calculated criteria, the different neutron spectra of the various irradiation locations must be considered. First, the spectrum at the inner edge of the vessel wall, Fig. 8, is noted to be quite different from the other spectra presented in this report. The effect of this difference is that, at the inner edge of the SM-1A reactor vessel wall, there is about 15 percent less calculated >1 Mev flux than fission spectrum > 1 Mev flux; conversely, there is about 35 percent more calculated > 0.5 Mev flux than fission spectrum >1 Mev flux. This is indicated in Fig. 17 by the square data points, which are placed about 10^{18} on the figure for convenience

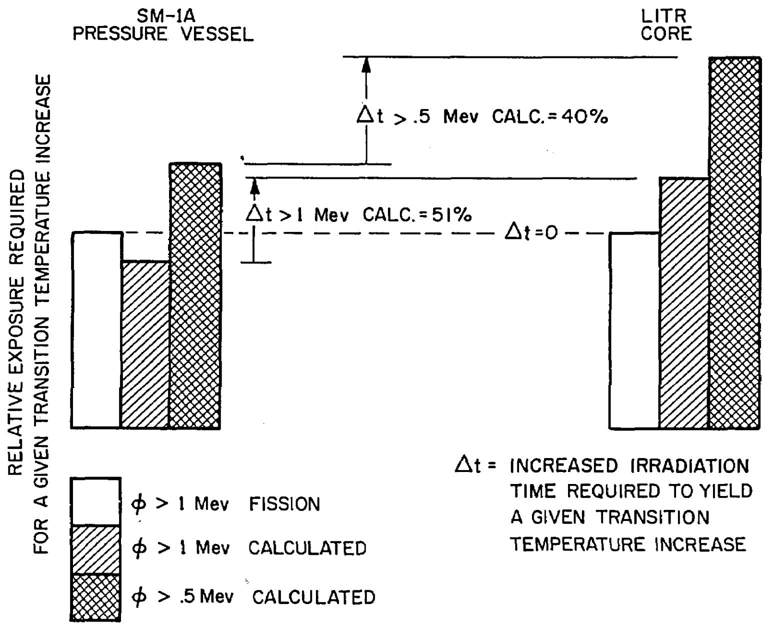

Fig. 18 - The relative exposure required in the SM-1A and LITR reactors for neutrons of three different criteria to effect a given transition temperature increase in A350-LF1 (Modified) steel. When the fission spectrum >1 Mev flux in both the SM-1A and LITR are the same, 51 percent more time will be required in the SM-1A to effect a given increase by calculated >1 Mev neutrons, and 40 percent more time in the SM-1A for calculated >0.5 Mev neutrons.

and do not relate to any particular time in the reactor lifetime; this concept is also shown graphically in Fig. 18. Furthermore, the LITR spectra indicate the presence of about 27 percent more calculated >1 Mev flux and about 89 percent more calculated >0.5 Mev flux than fission spectrum >1 Mev flux.

From Fig. 18 it can be seen, then, that when the instantaneous fission spectrum >1 Mev flux is assumed to be equal in both the SM-1A and LITR, the LITR flux is 51 percent more populous in calculated >1 Mev neutrons and is 40 percent more populous is calculated >0.5 Mev neutrons than is the SM-1A flux. Note that Fig. 18 presents the relative numbers of neutrons, as determined by the three exposure criteria, required to produce a given increase in transition temperature regardless of the reactor in which the increase is produced. It then follows directly (assuming an equivalent >1 Mev fission flux in both reactors) that 51 percent more exposure time will be required in the SM-1A to produce the calculated >1 Mev neutrons shown to be required by the LITR irradiations for a given transition temperature increase. Also, 40 percent more exposure time will be required in the SM-1A to produce the calculated >0.5 Mev neutrons shown to be required by the LITR irradiations for a given transition temperature increase.

In summary, the greatest benefit to lifetime would be accorded to the SM-1A reactor by using the calculated >1 Mev criterion, since it will require the SM-1A the longest additional time to produce the neutrons of this criterion necessary to effect a given transition temperature increase. Selection of the calculated >0.5 Mev criterion for the SM-1A reactor neutron exposure lifetime assessment might be more realistic, however, since this criterion encompasses a larger portion of the incident neutron spectrum. This criterion, as already noted, will not yield such an increase in lifetime and thus will add a note of conservatism to the analysis.

A safety factor such as this might be important, since the calculated spectrum-and-cross-section neutron exposure technique is in its infancy and depends greatly upon the accuracy of the representation of materials and components for computer analysis, as well as the accuracy of many nuclear physics constants required in the analysis. While the calculations for the purposes of this report were performed using the very latest and most authoritative information available, it should be recognized that research is continuing and refinements may be expected.

Finally, it should be noted that the fission spectrum >1 Mev criterion, as presently employed, will provide the most conservative value for embrittlement lifetime of the SM-1A reactor.

With the above analysis of steel behavior trends established, it would be necessary only to determine the actual flux at the SM-1A vessel wall, place it in terms of either of the calculated criteria or the fission spectrum >1 Mev criterion, and then extrapolate along the respective trend lines to determine a megawatt-year lifetime limit for the SM-1A vessel for a given transition temperature increase. It is not the purpose of this report to present such an analysis but only to provide the background information and procedures whereby it could be accomplished.

The foregoing analysis for projecting lifetime embrittlement for the SM-1A reactor pressure vessel based upon calculated spectra-and-cross-sections may be considered valid if the following assumptions are true.

1. The neutron spectra and activation cross sections determined by Dahl and Yoshikawa are accurate and are not significantly altered by minor core changes in the LITR reactor between different irradiations.

2. The significantly damaging neutrons in the reactor environments are fairly represented by either of the calculated exposure criteria.

3. There are no significant dose-rate effects for the A350-LF1 (Modified) steel in either the LITR core positions or in the SM-1A vessel wall.

The critical nature of transition temperature (NDT) increases in power reactor pressure vessels is sufficient justification to pursue every means possible to better understand how neutron spectra and embrittlement trends of pressure vessel steels can be best interrelated. It may well be that a relatively simple and basic technique, such as the determination of neutron spectra and adjustment of the flux monitor activation cross sections to encompass neutrons above a lower energy limit, 1 or 0.5 Mev, will prove to be quite satisfactory for the future assessment of pressure vessel lifetimes.

CONCLUSIONS

The metallurgical test specimen irradiations described in this report, wherein all variables were fixed except for the neutron spectral environment, have established a basis for intercomparisons of data from widely different exposure facilities. The results of this study to date indicate that data relating to the properties of steels irradiated in or near the core of light water moderated reactors can be confidently intercompared for engineering applications using the fission spectrum > 1 Mev criterion. This is possible because the magnitude of the shifts effected by the calculated criteria are on the order of the experimental data scatter. On the other hand, calculated spectra and average cross section adjustments to a lower energy limit > 0.5 or even as low as > 0.183 Mev must be applied to experimental data from highly moderated reactors in order to properly account for the total damaging-neutron exposure.

The water-steel environment of a light water moderated reactor pressure vessel wall is different enough from an in-core or near-core light water moderated test reactor environment, as evidenced by the SM-1A reactor study, that further research will be necessary to establish specific guidelines for engineering application of test reactor data to individual operating reactor pressure vessels. An important requirement in establishing such guidelines will be to obtain mechanical property data and neutron flux measurements directly at a pressure vessel for comparison with neutron exposures determined from calculated spectra-and-cross-sections. The behavior trends of pressure vessel steels resulting from accelerated test reactor irradiations can then be established for the different spectra and lower neutron energy limit criteria for comparisons with the actual vessel wall properties and measured dosimetry data.

It may be concluded from the results presented above that a significantly better understanding of the problem of neutron-spectral-radiation damage in pressure vessel embrittlement research can be obtained by placing all of the pertinent data in terms of a neutron exposure criterion which can eliminate major spectral differences.

Although some adjustments may be necessary in applying test reactor data to operating reactor cases, the sensitivity to nuclear radiation of the structural steels under study has proven that metallurgical factors appear to be of greater significance than adjustments for the damaging neutron spectrum. Until the mechanisms of steel sensitivity to irradiation are more clearly defined, and the means for their application yield steels of more consistent quality and response to irradiation, spectral considerations will probably continue to be a relatively minor contributor to the analysis and application of radiation effects data.

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13. ABSTRACT

Irradiation experiments to define behavior trends of reactor structural materials are frequently performed in reactor environments dissimilar to those expected during actual service. In order to accurately assess the damage produced in accelerated experimental environments so that the results can be applied to operating reactor cases, a damaging-neutron exposure criterion must be established which will account for the significantly damaging portion of the incident neutron spectra of both reactor environments. Several such exposure criteria have been evaluated through use of the results of metallurgical tests of reference steel specimens after irradiation in light and heavy water moderated reactor environments as well as in graphite moderated reactor environments.

The radiation-induced transition temperature or nil-ductility transition (NDT) temperature increases of the several steels involved are presented versus n/cm^2 determined by each of the following techniques: (a) assumption of a fission spectrum, extrapolation of activation data induced at a high Mev threshold to 1 Mev, and reporting exposure >1 Mev, and (b) calculation of spectra used to determine activation cross section for exposures above energy limits of 1, 0.5, and 0.183 Mev. The differences observed by this analysis were intercompared in relation to absolute magnitude as well as in terms of engineering significance. By applying these criteria to data relating directly to a pressurized light water power reactor, benefits to the lifetime of the reactor can be realized. The results of this study to date indicate that data relating to the properties of steels irradiated in or near the core of pressurized light water moderated reactors can be confidently intercompared for engineering applications assuming a fission spectrum and accounting for neutrons of energies >1 Mev. On the other hand, calculated spectra and average cross section adjustment to an energy limit as low as >0.183 Mev must be applied to data from highly moderated reactors.

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