Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY *(Corporate author)* Naval Research Laboratory Washington, D.C. 20390	2a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED 2b. GROUP

3. REPORT TITLE
MAJOR FACTORS AFFECTING NEUTRON IRRADIATION EMBRITTLEMENT OF PRESSURE-VESSEL STEELS AND WELDMENTS

4. DESCRIPTIVE NOTES *(Type of report and inclusive dates)*
Summary report; work on other phases of the work is continuing.

5. AUTHOR(S) *(First name, middle initial, last name)*
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6. REPORT DATE October 30, 1970	7a. TOTAL NO. OF PAGES 22	7b. NO. OF REFS 29

8a. CONTRACT OR GRANT NO. NRL Problem M01-14 b. PROJECT NO. RR 007-41-11-5409 c. AEC-AT(49-5)-2110 d. MIPR-ERG-11-69	9a. ORIGINATOR'S REPORT NUMBER(S) NRL REPORT 7176 9b. OTHER REPORT NO(S) *(Any other numbers that may be assigned this report)*

10. DISTRIBUTION STATEMENT
This document has been approved for public release and sale; its distribution is unlimited.

11. SUPPLEMENTARY NOTES Also sponsored by the U.S. Atomic Energy Commission	12. SPONSORING MILITARY ACTIVITY Dept. of the Navy (Office of Naval Research) Arlington, Va. 22217; Dept. of the Army (Nuclear Power Field Office)

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14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Neutron irradiation embrittlement						
Pressure-vessel steels						
Fracture toughness						
Nuclear vessel						
Composition						
Weldments						
Weld heat affected zone						
Neutron fluence						
Fracture analysis						
Radiation damage surveillance						
Material specification						
Controlling embrittlement						

CONTENTS

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PROBLEM STATUS

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AUTHORIZATION

NRL Problem M01-14
Projects RR 007-41-11-5409 and AEC-AT(49-5)-2110
MIPR-ERG-11-69

Manuscript submitted August 7, 1970.

MAJOR FACTORS AFFECTING NEUTRON IRRADIATION EMBRITTLEMENT OF PRESSURE-VESSEL STEELS AND WELDMENTS

INTRODUCTION

This report reviews in summary the influence of several major variables involved in the process of pressure-vessel steel embrittlement by neutron radiation. Results have been drawn from programs sponsored by the U.S. Atomic Energy Commission (AEC) at NRL and at other laboratories which are aimed at developing the knowledge to assure structural reliability of large commercial nuclear reactor pressure vessels. Advances within these AEC programs provide the basis for a critical review of the several aspects of steel embrittlement.

Portions of large commercial nuclear reactor pressure vessels may be exposed to significant levels of high-energy neutrons during service life. The most critical region for such bombardment is the midcore region. This is fortunate in that structural complexities or discontinuities and hence local regions of high stress are minimized in this region; however, most reactor vessels are made up of welded plates which provide the opportunity for some small metallurgical, and possibly physical, discontinuities. Thus, consideration of the physical construction aspects of a reactor vessel must be coupled with knowledge of the nuclear environment, the temperature, and the gradation of materials properties through the reactor vessel wall in the midcore region. This report examines the latter, emphasizing especially the several factors which influence radiation embrittlement, considering their roles, their interrelationships, and ways in which they can be turned to minimize such effects.

The irradiation condition of a reactor vessel is a moving target throughout its life. Hence, considerable effort must be exerted to assure as much knowledge as possible of the variable factors which influence embrittlement. The most important factors are the materials and their response to irradiation as well as the nuclear and physical environments involved, especially the vessel temperature at the position of highest neutron exposure. Within the materials variability factor are the differences between plate, weld, and weld heat affected zone, variations due to thickness and metallurgical orientation of grain structure due to the plate formation practice, and the possibility of physical or metallurgical discontinuities associated with the welding process. The question of nuclear environmental factors, their definition and especially the possibilities of flux rate and neutron spectrum effects, which are not easily projected from early data in a given plant, must also be considered in order to interpret or project the condition of a reactor vessel. The temperature, which in more recent reactor vessels is progressively higher, also has a major effect and in most cases corrects or minimizes radiation embrittlement.

These factors, their relationships, and their potential consequences are reviewed. This is done within a framework of evolving fracture analysis procedures having direct applicability to the steels normally used for commercial nuclear vessel construction. Areas requiring additional study are reviewed along with conclusions as to where metallurgists stand now regarding this question.

ASSESSING RADIATION EMBRITTLEMENT

In procurement, reactor vessel steels are required to meet certain mechanical properties limitations. Strength and notch ductility are included. These requirements only assure a reliable vessel at reactor startup. From that time forward the properties of a vessel exposed to significant numbers of high-energy neutrons change. The extent of change depends on many things, but a most difficult aspect of this question is the "moving target." Many studies of radiation effects in pressure-vessel steels have been conducted, and the most significant effects observed from a vessel reliability point of view are those which reduce ductility; hence, the emphasis here.

Two distinct but interrelated aspects of embrittlement are definable. First, there is the very visible increase in the ductile-to-brittle transition temperature with neutron exposure, and, second, the less predictable but nevertheless important reduction in fracture resistance as measured by the energy absorbed in a dynamic fracture test. Historically the Charpy-V impact test (1) has been the primary basis for specification limitations and for defining radiation effects on notch ductility. The significance of Charpy-V data has been complemented through comparisons with the nil ductility transition (NDT) temperature as determined by the drop weight test (2) and, more recently, through a more comprehensive approach to dynamic fracture evaluation, the NRL Dynamic Tear Test (3). In addition, in recent years, the compact tension specimen (CTS) has been applied in an attempt to provide a more quantitative approach to fracture analysis (4). As might be expected, there have been progressive clarifications of certain aspects of fracture with this evolutionary process.

Fracture Analysis Concepts

The increase in transition temperature is usually indexed and a trend established for the increase (ΔT) as a function of the neutron fluence (n/cm^2) greater than some energy threshold, usually 1 MeV. This trend can then be used as an operating guide to assure that the vessel is always in a condition of maximum ductility while significant operating stresses are applied. The reduction of fracture resistance, or, as it is often described for the Charpy-V notch test, the "shelf drop," is not so readily amenable to an operating-engineering solution since the state of knowledge of the critical point for low-energy tearing, the point where energy absorption becomes marginal for assuring structural reliability, is not as well developed. Nevertheless, two developments give aid in this area. First, Pellini (5) has developed a concept for classifying materials by their inherent fracture toughness characteristics. This is done through the Ratio Analysis Diagram (RAD) approach which permits a semiquantitative indexing of fracture toughness through comparisons of the dynamic tear or Charpy-V notch energy and the plane-strain stress intensity factor at the onset of unstable fracture, K_{Ic}, versus the yield strength. Placement of such data on the RAD produces guidance as to its relative resistance to fracture. Radiation-induced reductions in fracture resistance can thus be indexed and critical energy absorption levels identified. This identification depends on the knowledge of both irradiated fracture and strength characteristics.

Hawthorne (6) has made several initial RAD analyses on irradiated A302-B, A533-B, and A543 steels. His initial results agreed well with the work of Barsom and Rolfe (7), who have developed an empirical correlation between dyanamic K_{Ic} and Charpy-V (fatigue cracked) shelf energy data (CVN). This correlation, for several high-strength steels (levels about the same as the irradiated steels), produced the relationship

$$\left(\frac{K_{Ic}}{\sigma_y}\right)^2 = \frac{5}{\sigma_y}\left(CVN - \frac{\sigma_y}{20}\right)$$

Continued developments in this area should permit the establishment of a shelf energy limit below which the operational *stress-temperature* limitations would be superseded by operating *stress* limitations to assure vessel structural integrity. It is beyond the scope of this report to describe in detail the fracture analysis aspects. However, results involving the four tests mentioned will be used as appropriate for describing selected aspects of the subject.

Physical specimen size limitations for irradiation studies have made it impossible to fully evaluate or even closely simulate the notch ductility of a vessel as it is used. Accordingly, smaller specimens must be used to project the total fracture toughness of a reactor vessel. Extrapolation inherent in this process is now believed to be realistic in view of the results (8) obtained using dynamic tear specimens of full thickness (12 inches) in comparison with small-specimen results. Results on compact-tension specimens to provide K_{Ic} values (4) for full-thickness (12 inches) A533-B steel have also aided confidence in small specimen projections. (For more detailed reading relative to pertinent fracture analysis concepts, see Refs. 5 through 12.)

Another highly significant aspect of the full-thickness fracture tests was the definition of a region for valid application of linear elastic fracture approaches and the region where such validity would not be expected, indicating an upper limit for valid static K_{Ic} about at the midtransition range as defined by the large dynamic tear tests. In addition, the full-section dynamic tear tests described a thickness-induced mechanical constraint with an elevated and sharpened transition region approximately 70°F (39°C) above that described by thin-section tests. Accordingly, the projection of fracture toughness as defined by small dynamic tear or Charpy-V tests requires interpretation in a conservative way (such as superimposition of the 70°F to the accepted NDT + 60°F value) for projecting the reliability of thick structures. These fracture analysis aspects of the problem are extremely important and must be considered in establishing or projecting irradiated vessel conditions.

Embrittlement Assessment

Two approaches to the assessment of radiation embrittlement of reactor vessel steels have been used. The first involved planned laboratory studies of typical commercial and specially procured laboratory or commercial-scale heats of the vessel steels, and, second, the evaluation using surveillance specimens of a particular reactor vessel exposed in that reactor at the vessel wall to provide data on the exact changes occurring. These two approaches have been found to be complementary and without significant anomalous results attributable to the very different rates of irradiation involved.

Irradiation of pressure-vessel steels at temperatures representative of commercial water reactor vessel service conditions has indicated significant changes in strength and ductility. The changes in notch ductility have been cataloged over a period of years; hence, the role of individual variables can be assessed. The approaches which have been used in assessing radiation embrittlement to date and the data produced provide the basis for defining the critical factors in neutron embrittlement. Thereby, these approaches guide the operational or design decisions which will assure continued operational reliability for plants in service and will minimize concern for the reliability of containment systems of plants yet to be placed in service. The basis for this statement and the nature, extent, and interaction (sometimes competing) of various factors affecting the degree of radiation embrittlement of vessel steels are reviewed on the following pages.

CRITICAL ASPECTS OF NEUTRON IRRADIATION EMBRITTLEMENT

Neutron Environment

The first step toward a projection of the degree of embrittlement for a given reactor vessel involves the measurement of the neutron flux at the vessel wall for comparison with steel embrittlement data from surveillance or experimental studies. In an operating nuclear plant this is usually accomplished through the placement of specimens of the vessel steel and neutron dosimeters of the activation threshold type at the vessel wall for periodic removal and evaluation. After the initial surveillance step, it is possible to project (assuming certain facts relative to future reactor operation) exposure levels and embrittlement in future years.

A three-step procedure is the most desirable approach to defining the neutron environment: (a) the measurement of the flux, (b) the computation of the spectrum at the vessel wall, and (c) the projection of the spectrum in terms of its damaging potential through a weighting process for that particular spectrum.

The measurement of neutron fluxes is not simple and, in fact, involves procedures not normally carried out as thoroughly as is desirable to assure full knowledge of the nuclear environment at a reactor vessel. The usual procedure involves the selection and exposure of a limited number of elements (for practical reasons usually metals as foils or wires) which are activated by exposure to neutrons. This activation, which is then evaluated in terms of decay radiation, provides a measure of the number and types of neutrons impinging on the reactor vessel.

A usual basic neutron dosimetry package involves bare cobalt for thermal neutrons, cadmium-shielded cobalt for epithermal neutrons, and iron for the high-energy neutrons. This is really a skeleton on which a more complete and definitive analysis might be built but which, with experience, has provided key information for projecting neutron embrittlement of reactor steels. However, this skeleton is quite limited since the thermal and epithermal neutrons are not very effective in causing damage in steels. Similarly, the iron detector is the most effective in describing the higher energy ranges. In fact, one measurement (13) shows that at a neutron energy of 2.3 MeV the cross sections* for the reaction $^{54}Fe(n,p)^{54}Mn$ was only 26 mb, whereas the peak cross section, ≈ 470 mb, is reached at about 5.5 MeV. Thus, highly significant portions of the neutrons between the epithermal region (keV) and the high-energy region (above 2 MeV) are not well defined using this dosimetry approach. Recently, however, the trend has been towards the use of additional dosimeters in experiments if not in surveillance. The addition of certain fission detectors and other threshold detectors has provided broader coverage (14) of the neutron spectrum and thereby complements the neutron spectrum calculation which is the second step in the procedure.

Measurement of the neutron spectrum is not critical where projections of vessel embrittlement are based solely on surveillance. However, in order to make the best projections, it is necessary to make comparisons with experimental data from test reactors covering a range of fluences or at least to compare data from accelerated exposure (nearer the core) positions in the same reactor. Also, if projections to future years are to be made, good dosimetry for each surveillance period is essential in order that changes in core loading or power level may be reconciled. Differences in the spectrum from the test reactor to power reactor or from accelerated to vessel wall position must be taken into account and adjustments made. For example, in the Big

*The propensity for the production of the reaction expressed in the usual terms of cross-section measurements—millibarns (mb).

Rock Point vessel surveillance, both accelerated and vessel wall specimens were available. Figure 1 (15) describes the computed spectra for these two locations plus that for the interface point between the A302-B vessel steel and the stainless-steel cladding. Note the rather large discrepancy in shape above about 0.5 MeV. Such differences require adjustment. An insert to Fig. 1 depicts the relative flux of neutrons (>0.5 MeV) for several locations from the core to the outside of the vessel wall. Acceptance of such a distribution depends on careful spectrum analysis. (Note the severe drop in flux through the vessel wall. This drop indicates the conservatism inherent in accepting inner vessel wall data for projecting embrittlement.)

Fortunately, advances in neutron spectrum calculations now make it possible to readily reconcile differences between spectra. As a generalization, however, experience involving different spectra in water-cooled test reactors and water-cooled power reactors suggests that required adjustments for the variations in spectrum result in relatively minor changes in projected embrittlement. For example, in a recent study at NRL, comparisons were made between a fission spectrum, which is often the assumed basis for comparisons, and the actual spectrum in a test reactor. In this case a 20% downward adjustment from the fission spectrum fluence to the actual spectrum fluence resulted. Such an adjustment would not be critical except where embrittlement is very great. Nevertheless, with the techniques for the calculation of the spectrum and for dosimetry being improved, fuller use of these improvements should be made in order that early projection of vessel embrittlement might be made with greatest confidence.

The third area, that of damage function analysis, is in relative infancy compared with neutron dosimetry and spectrum measurement techniques. Nevertheless, this approach will permit refinements in projection to the future. This procedure involves weighting the calculated neutron spectrum in terms of its damaging contribution by neutron energy group. This approach was developed and has been tested using vessel steel embrittlement data (16). Advances in this area should soon develop results from which to select a preferred threshold (>? MeV) for describing the damaging neutron exposure at a reactor vessel.

Other Environmental Factors

Temperature — The irradiation or service temperature of vessel steels is very important. Higher temperatures generally result in reduced embrittlement within the range of 400° to 600°F (204° to 316°C) which is common to water reactors. This is attributed to the healing effects of higher temperature in providing the energy to cause displaced or interstitial atoms to move to vacancy sites or to other relatively innocuous locations rather than aggregating to affect bulk properties.

The very significant influence of irradiation temperature is illustrated in Fig. 2 in which comparisons are made for a reference heat of A302-B steel exposed at <450°F (232°C) and at 550°F (288°C). These data represent a compilation from many NRL experiments and clearly demonstrate the effect of increased temperature as well as the uniformity achieved when one steel is irradiated to different fluences at one temperature. The advantage of an even higher temperature, 585°F (307°C), in the relatively low flux levels of an operating power reactor, the Big Rock Point Reactor, is shown by the single point at a fluence of 7.1×10^{19} n/cm^2 > 1 MeV (15).

If the exposure temperature is even higher (e.g., 650° or 750°F, 343° or 399°C), progressively lower embrittlement is usually achieved. However, higher-alloy steels, such as 3-1/2Ni-Cr-Mo steel (similar to A543), may be further embrittled through complementary and sometimes radiation-enhanced, thermal aging or metastability phenomena. Nevertheless, for the steels now in service, higher temperatures (to 750°F, ≈400°C) result in lower embrittlement. This observation is the basis for the use of higher than operating temperature for annealing (correcting) vessel embrittlement.

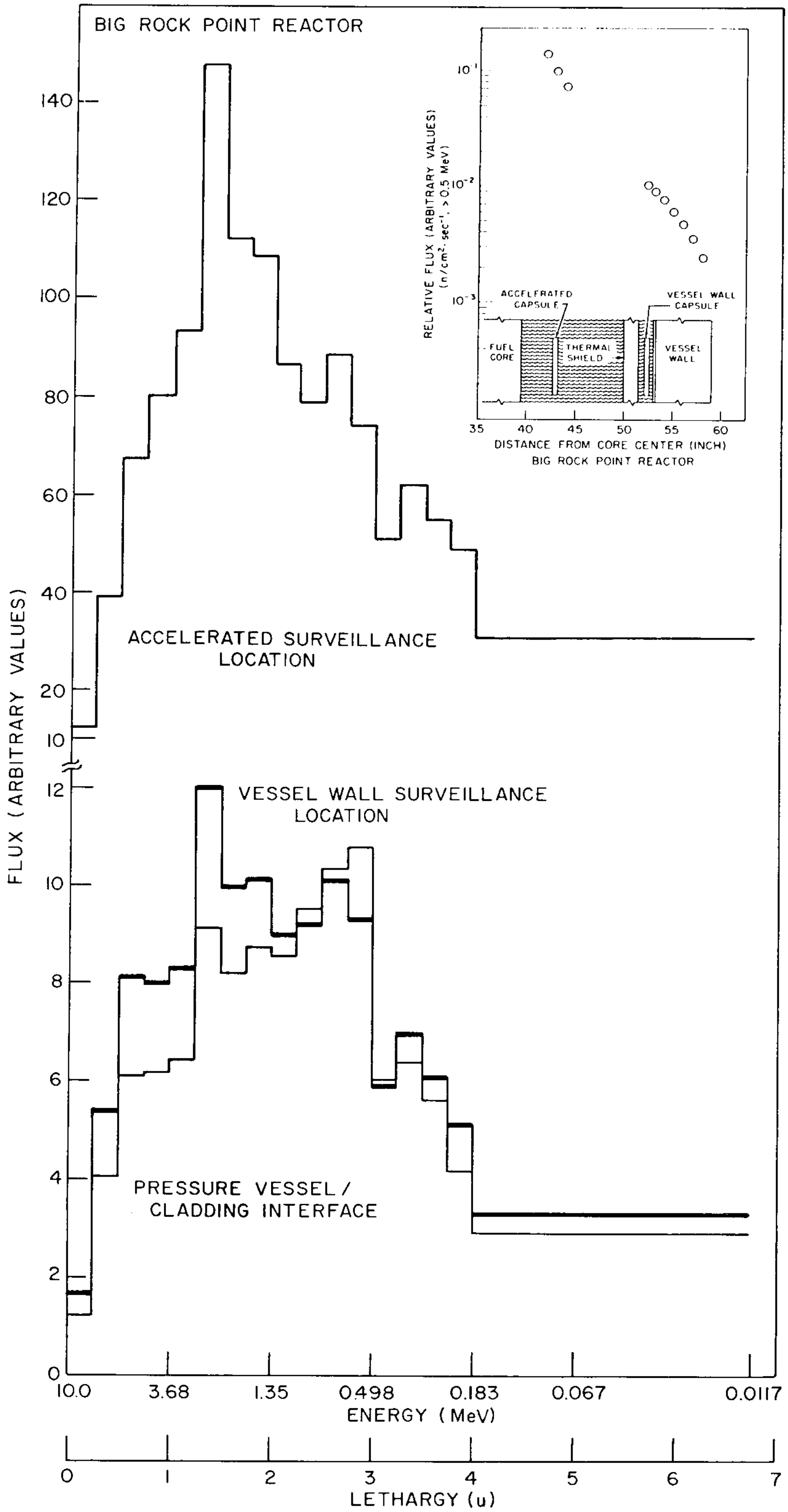

Fig. 1 - Graphical representation of the neutron spectra at the accelerated and vessel wall surveillance positions as well as at the pressure vessel-stainless steel cladding-interface position in the Big Rock Point Reactor (BRPR). Flux values are in arbitrary units. The spectra show a shift in intensity toward lower energies for locations farther from the fuel core. The inset at the upper right provides a schematic representation of the decreasing neutron flux (> 0.5 MeV) from the fuel core edge through the vessel wall.

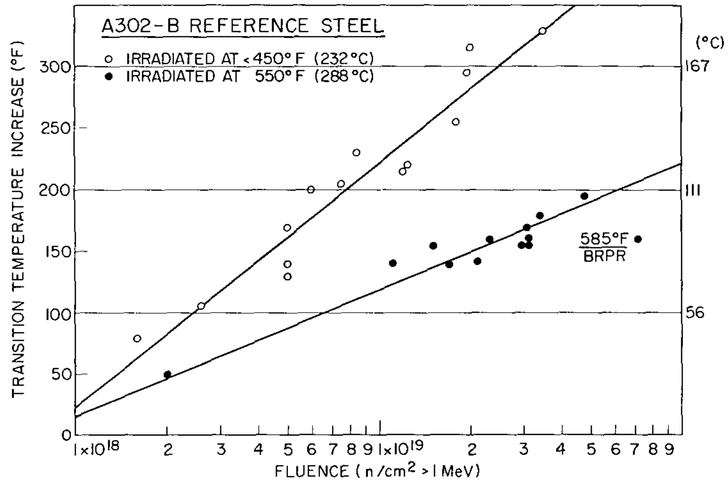

Fig. 2 - Effects of temperature on the radiation-induced increase in transition temperature for a reference heat of A302-B steel

Applied Stress — The effect of applied stress on the radiation embrittlement of pressure-vessel steels has been considered in limited experiments. In two experiments carried out in the USA (17,18), no significant effect of irradiation under stress versus the unstressed condition was observed on A302-B steel irradiated at stresses of 20 and 80 percent of yield strength, respectively. The results of these experiments suggest that concern for the interaction of applied stress and radiation may be effectively ignored so far as embrittlement of current pressure-vessel steels is concerned. It is obvious, however, that the effect on flaws and flaw growth must not be ignored in considering the total reliability question for reactor pressure containment systems.

Materials

The variable affecting the level of embrittlement which offers the greatest potential for control is materials since large differences have been observed and the level of embrittlement has been shown to be controllable under certain conditions. Practically, however, since a large number of vessels for water reactors have already been constructed or the steels produced, the process of selecting or producing steels of low-radiation embrittlement sensitivity using criteria developed in laboratory studies is limited to those vessels for future reactors. Nevertheless, such criteria or guidelines are valuable for judging the potential embrittlement of any vessel steel.

Early indications of variation in sensitivity to radiation were observed in experiments comparing a number of then current or potential vessel materials. For example, irradiation in one NRL experiment (19) carried out at 550°F (288°C) to a neutron fluence of 3.8×10^{19} n/cm^2 > 1MeV produced varying degrees of embrittlement ranging from a ΔT of 185°F (103°C) for A212-B pressure-vessel steel to 90°F (50°C) for A543 to 15°F (8°C) for an experimental 7-1/2Ni-Cr-Mo steel. Such wide variations added impetus to the study of possible variations among heats of the same steel. Accordingly, studies were made of a number of heats of A302-B and A533-B steels. In one study (20) on ten different heats of A302-B steel, it was possible to identify two distinct trends, low and high sensitivity. The difference between the two was approximately a factor of two with the worst of the sensitive group yielding serious embrittlement implications. Similar results have been obtained on commercial A533-B pressure-vessel steels. Comparison of the results (Fig. 3) from a number of NRL experiments indicates a similar, two-group division of the poorest and the best.

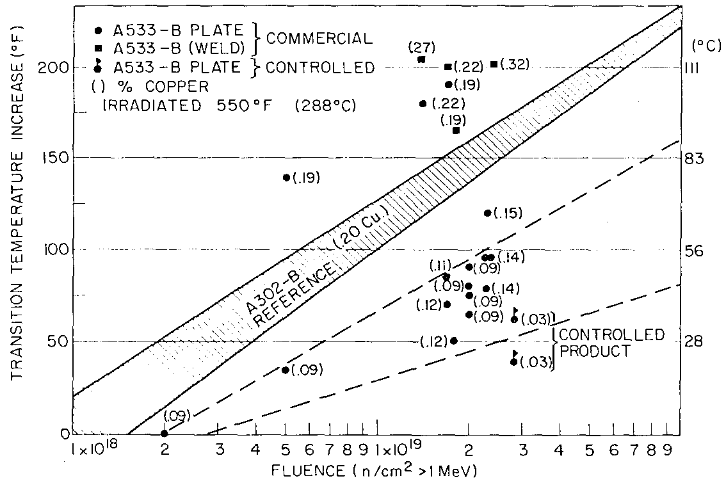

Fig. 3 - Radiation-induced increases in the transition temperature of commercial steel plates and weldments of type A533-B irradiated at 550°F (288°C) to various fluences. The copper content is presented beside each point. Steels containing more than 0.19% copper exhibit an exaggerated embrittlement, whereas those containing less than 0.14% copper consistently show less embrittlement than the A302-B reference steel (central band). All of the commercial-scale welds studied to date fall in the "sensitive" group. Data for a controlled specification heat having very low copper (0.03%) fall in the lower portion of the "insensitive" group.

Some investigators have categorized radiation-induced transition increases in terms of $\Delta T = A(\phi t)^{1/2}$ or $A(\phi t)^{1/3}$, where A is a constant and ϕt equals the neutron fluence above some threshold. It is clear that no such equation can adequately describe the data for A533-B steels because of the wide divergence of the data.

As a result of a number of NRL laboratory studies on specially fabricated (laboratory-scale) A302-B steel, it has been possible to identify elements critical to the embrittlement process, especially copper and phosphorus. Through control of these and other residual elements it has been possible to consistently produce pressure-vessel steels on a laboratory scale having high resistance to radiation. Accordingly, in comparing the two groups, sensitive and insensitive A533-B class 1 (Fig. 3), the criterion of copper content was applied (phosphorus variations were relatively small). Values for copper content are provided beside each point. In addition to the data on A533-B steel, for comparison purposes, a volume of data for a reference A302-B composition containing 0.20% copper has been used to provide the central trend band of Fig. 3.

In order to evaluate the possibilities for producing a controlled, commercial-scale product, having high radiation resistance, the AEC authorized the procurement by NRL of a 30-ton heat in which the residual-element contents were controlled. Limitations were imposed especially on copper (0.03%) and phosphorus (0.009%) with a combined limitation on arsenic, antimony, tin, and bismuth (0.05%). The results are indicated for comparison on Fig. 3 in the bracketed points at the lower right (21). Of the A533 steels studied the level of copper appears to determine the position or group within which a steel falls. The group falling above the A302-B trend band contained copper at levels between 0.19% and 0.32%. The radiation-resistant or insensitive group of commercial A533 steels contains copper levels between 0.09% and 0.14%.

Fracture resistance is measured both by the degree of upward shift in NDT or ΔT and by the reduction in energy absorption or shelf drop. The former is indicated by the data plotted in Fig. 3. The shelf drop has been defined by Burghard and Norris (22) for older irradiated steels in terms of a $\Delta E/E$ (shelf drop over initial shelf level in ft-lb) trend line equal to

$$-5.70143 + 0.145 \log \phi t,$$

where ϕt equals the neutron fluence >1 MeV. Newer steels studied at NRL described a much lower slope than this equation indicates, similar to the trend ($\Delta E/E = -2.63287 + 0.063 \log \phi t$) described by Kawasaki et al. (23). In the case of the controlled composition heat the ΔE was extremely small in spite of the fact that the initial E was about 140 ft-lb. Unfortunately, even the newer welds of A533-B often exhibit $\Delta E/E$ values falling in the higher trend described by Burghard. This factor coupled with low initial E and greater ΔT values raises additional concern for weld quality after irradiation (24).

The highly successful demonstration of the capacity for producing, on a commercial scale, radiation-resistant steels provides an opportunity and a challenge to the industry. As the best of the commercial steels already approach the results for the controlled product, with proper composition specifications and care in steelmaking, it should be possible to produce vessel steels having high resistance to radiation embrittlement and thereby to minimize concern for this problem in future vessels.* Unfortunately, examination of Fig. 3 indicates that all the weldments studied are sensitive to radiation. Thus, success in producing plate steel having high resistance to radiation must be followed by success in producing welds of similar high resistance. Such work, now underway at NRL, has shown early indications of success.

To test the role of copper in the embrittlement process, the controlled heat of A533 was split so as to produce a high-copper version, in this case 0.13%. This produced an embrittlement more than twice as great as that for the 0.03% copper version. Since all other components were unchanged, this verifies again the important role of copper in the embrittlement at 550°F (288°C) for this particular pressure-vessel steel. Experiments are being carried out to determine the exact role of copper in the embrittlement process. Three approaches to this problem are being taken: (a) to examine the possibility of diffusion and segregation of copper, (b) to examine the role of copper as a sink for radiation-produced defects, and (c) to observe, using the electron microscope and other techniques, the nature and distribution of radiation-defect aggregates in copper-doped and pure-iron foils.

In summary, it would appear that for the current compositions of pressure-vessel steels it is possible, through composition specification and care in steelmaking practice, to produce a product which is essentially immune under the normal conditions of nuclear service to serious radiation embrittlement.

Similar success in controlling the level of embrittlement in a higher-strength, potential pressure-vessel steel (type A543 steel and its weldment) has also been demonstrated. Plates of several commercial A543 steel heats have shown high radiation resistance, but welds of this steel have shown very high sensitivity, one weld exhibiting a shift of 535°F (297°C) after 3.5×10^{19} n/cm^2 > 1 MeV at 550°F (288°C) (14). This commercial weld contained 0.42% copper. A controlled-copper (0.02%) weld shifted only

*Sterne (25) of Lukens Steel has compared the relative levels of copper, phosphorus, and sulfur in heavy-section heats of A302-B steels produced between 1957 and 1960 with those of A533 steel produced in 1968. Significant reductions in all three elements have been produced in later production. For example, copper ranged from 0.09% to 0.17% in 1968 versus 0.11% to 0.27% in 1957-1960.

45°F (25°C) under the same irradiation conditions and retained the high initial energy absorption capacity at shelf level temperature. This work is continuing with the hope of providing weld composition specification limitations which will be used to produce a total reactor vessel that is essentially immune to neutron embrittlement.

SUMMARY AND CONCLUSIONS

Space does not permit a full discussion of the many aspects of radiation embrittlement which must be considered in evaluating pressure-vessel reliability. However, the more important of these are reviewed briefly here along with conclusions relative to the major points discussed above. In addition, areas requiring additional research or clarification are cited.

Neutron Environmental Factors

Three aspects of the neutron environment are identified.

1. Neutron Flux. The rate of irradiation varies significantly between test and power reactors. Because of this variation, studies have been advanced to evaluate the possible effects of flux rate. Results to date suggest no significant effects over a range of approximately one order of magnitude difference.

2. Neutron Spectrum. The neutron energy spectrum varies from place to place in a reactor and from one reactor to another. Compared with other types of reactors, light-water-moderated reactors exhibit relatively smaller variations in the neutron spectrum. Accordingly, though adjustments are necessary for making projections for service locations different than the irradiation location, these are usually relatively small. Nevertheless, techniques are available to permit the computation of a spectrum for a given location in a reactor, thereby permitting a better projection of future embrittlement.

3. Damage Potential. The damage potential or damage function for any spectrum depends on the shape of that spectrum, the material exposed, and the property being evaluated. For steel embrittlement it has been shown that high-energy neutrons, especially those above 0.1 MeV, account for essentially all of the damage in most of the spectra to be found in a light-water-moderated reactor (26). Refinements in damage function analysis are now underway to further aid in evaluating this factor.

Surveillance Aspects of the Neutron Environment

In an operating nuclear plant with a good surveillance program the effort applied to neutron environment definition may be minimized. This is true because specimens at the vessel wall represent the vessel very well. Projections of properties changes can be made from such specimens. Nevertheless, the ideal surveillance program has not been the practice. In other words, surveillance programs are usually kept small for practical reasons. Accordingly, projections to the end of life may be difficult based on specimen surveillance alone. Accordingly, measurement of the neutron environment for the peak flux regions* of the vessel provides important guidance for predicting vessel embrittlement. It should be noted that early projections of lifetime fluences from computations in such reactors as the Dresden, the Big Rock Point, and Yankee-Rowe

*The inside diameter of a water reactor vessel is as near circular as it is possible to make it but the fuel core is not a circle; hence, points or regions of peak flux can be expected.

were low. In the case of the Big Rock Point, computations were low by an order of magnitude compared with projections based on measurements in the plant. In spite of weaknesses recognized in the past, however, surveillance programs with good neutron dosimetry provide the basis for realistic projections of future embrittlement.

Past experience, both from surveillance and experimental efforts, provides important guidance and permits the following generalization. If the expected vessel lifetime neutron fluence is less than 10^{19} and the reactor operating temperature is significantly above 500°F (260°C) for most of that lifetime, concern for vessel embrittlement is minimal regardless of the particular vessel steel.

Other Environmental Factors

1. Temperature. The exposure or service temperature greatly affects the degree of embrittlement. Irradiation at 450°F (232°C) often results in embrittlement twice as great as that observed at 550°F (288°C). The healing effects of higher temperature offer a technique for correcting radiation damage, that is, through annealing at a temperature significantly above the normal service temperature. Annealing has been carried out successfully in the Army SM-1A reactor. Thus, the progressive trend toward higher operating temperature with newer plants tends to minimize the anticipated neutron embrittlement. (It should be noted, however, that (a) those steels defined as sensitive to embrittlement exhibit the sensitivity especially at elevated temperatures and (b) certain higher-alloy steels have exhibited a combined temperature plus radiation embrittlement which exceeds that produced by either thermal aging or radiation alone.)

2. Applied Stress. Several experiments to assess the combined effects of radiation and applied stress have identified no significant effect of the applied stress during irradiation. Since higher-alloy steels are used, the strain aging effects which may come into play, however, make this factor more critical.

3. Fatigue. The fatigue characteristics of pressure-vessel steels have shown no significant effect of irradiation in studies conducted to date. However, it should be noted that these studies have essentially involved initiation and fatigue life studies rather than the more important fatigue crack propagation rate studies. The latter are now being conducted on irradiated steels.

4. Thermal Aging. The thermal aging factor has been tested through test reactor experiments and through the placement of control specimens out of the radiation region in operating power reactors. For the currently used pressure-vessel steels no significant thermal embrittlement effect apart from or supplementary to radiation embrittlement effects has been observed. With higher-alloy steels now being considered for nuclear service, this is not the case and thermal aging phenomena accentuated by irradiation have been observed. This factor will demand more attention with the application of progressively higher-alloy, higher-strength pressure-vessel steels.

5. Hydrogen Embrittlement. The possibilities of hydrogen production through corrosion mechanisms or of the presence of hydrogen from steel production have been considered and its embrittling effects assessed with negative results on currently used reactor steels (27-29). However, as higher-strength steels are applied in more severe environments, this factor may become more significant and require further study.

Materials

1. Steel-to-Steel Variability. Large variations have been observed in embrittlement as defined by ΔNDT or ΔT (Charpy-V) from one composition to another. These range to as much as an order of magnitude for a given irradiation condition. Both the microstructure and the composition (especially the latter) are effective in producing or accentuating wide variations.

2. Heat-to-Heat Variability (Same Steel). Again, very large variations (to an order of magnitude difference in an A543 steel weldment) have been observed, in spite of the same nominal composition. The importance of this factor cannot be overestimated in any conservative attempt to fully understand or to predict the radiation embrittlement of a reactor vessel.

3. Plate Versus Weld Versus Heat Affected Zone. For the current pressure-vessel steels, the heat affected zone has consistently indicated low sensitivity to radiation embrittlement while welds have routinely indicated higher sensitivity than the other two components (see Fig. 3). The embrittlement variations among plates of recent commercial type A533-B steels have been very wide, ranging to differences as large as a factor of three with the related weld metals falling predominantly on the upper end of the embrittlement scale.

4. Higher-Strength Steels. Similar variations in radiation embrittlement sensitivity have been observed on higher-strength steel plate and weld metal, especially on the A543 weldments. In general, however, commercial plate products of the higher-strength steels have generally shown lower embrittlement than the similar products of type A302-B or A533-B though a thermal metastability has been observed which is accentuated by radiation exposure at elevated temperatures. Studies are continuing to examine full commercial-scale products of the higher-strength steels having controlled compositions.

5. Specimen Orientation and Fracture Resistance. The effect of orienting specimens either parallel or transverse to the primary rolling direction has resulted in significant variations in fracture resistance before and after irradiation. If the cross-rolling ratio in plate production is high and transverse properties are significantly inferior to the parallel or longitudinal properties, the low initial fracture resistance or energy absorption characteristics may not allow a significant radiation-induced reduction without producing concern for low-energy tearing at any temperature. Fortunately, the steels showing higher resistance to radiation embrittlement also have higher initial fracture resistance or shelf toughness in dynamic loading and exhibit lower reduction due to radiation. Unfortunately, the reverse is also true.

6. Thickness Aspects. Three factors—fracture behavior, properties gradients, and neutron attenuation—are involved in the thickness aspects. On an unirradiated basis full-section dynamic tear (DT) tests indicated a transition (sharper but later) about 120° to 150°F (49°-66°C) above NDT, thus validating thin-section test results relative to the dramatic rise in fracture resistance but with a thickness effect of ≈70°F (39°C) from the thin to thick section tests. Neutron attenuation results in progressively smaller increases in NDT, but properties gradients (higher than NDT in the central portion of a thick plate) nullify at least a part of the advantage resulting from reduced embrittlement because of through-thickness neutron attenuation. Nevertheless, the attenuation factor results in a high degree of conservatism relative to full-vessel embrittlement when the inside wall condition is taken as the reference point for embrittlement projection.

7. Tensile Properties. In general, the current pressure-vessel steels under typical or even severe nuclear service conditions have exhibited a high degree of retained tensile ductility in spite of very significant increases in yield strength. Under severe irradiation conditions, however, the yield to ultimate strength may become very low, indicating little additional capacity for work hardening or for yielding to alleviate high local stresses.

Another important aspect which should be noted is the pattern of yield strength increases versus Charpy-V energy shelf reduction. In the first stage of radiation embrittlement yield strength increases relatively slowly while the shelf drops significantly. In a second phase, the shelf drops considerably less while yield stress increases significantly, and in a third stage, the shelf drops significantly with no significant additional increase in yield strength. These observations suggest the need for examination of radiation embrittlement to very high neutron fluences to test this phenomenon. The most significant aspect of this pattern lies with the possibility of reducing fracture resistance with increasing strength to a point where higher temperatures no longer provide sufficient ductility to avoid low-energy tearing or even plane strain elastic fracture in thick sections.

Controlling Embrittlement Through Materials Specification

Through control of composition, especially that of the residual elements copper and phosphorus, it is possible to produce steels having consistently high-irradiation embrittlement resistance. Copper contents in the range of 0.08% to 0.14% generally indicate steels of relatively low sensitivity, while levels greater than 0.19% or 0.20% are associated with high sensitivity. Superior radiation resistance has been demonstrated in a controlled-composition 30-ton heat of A533 steel containing 0.03% copper and 0.009% phosphorus. For a given level of copper and phosphorus, the cast weld structure appears to be more sensitive to embrittlement, however. Hence, the critical component for controlling embrittlement in a full reactor vessel appears to be the weld, and welds have shown superior radiation resistance when composition, particularly copper, is controlled.

However, examination of a commercial weld in three locations (weld root, midthickness and upper-level, 1/4T from top surface) indicated differences in sensitivity paralleling differences in copper content. Thus, weld production as well as sampling for surveillance must be carefully carried out. For surveillance, care is required to insure the selection of the most representative weld material. This proved to be the material from the middle and upper portions of the weld which contained higher copper and showed greater sensitivity to radiation. This material represents approximately three quarters of the weld thickness; hence, the sampling problem only requires that the weld "root" region be avoided as it is often diluted more with the base plate material and thus may be lower in copper than the bulk of the weld. The ultimate solution appears to be in assuring no higher (and preferably lower) copper levels in the weld wire than in the base plate.

Techniques for minimizing radiation embrittlement of future vessel steels have been reviewed. By integrating these concepts and applying conservative fracture analysis approaches, it should be possible to minimize the concern for the reliability of vessels constructed in the past as well. Nevertheless, a continued alertness and strong efforts to gather data on each reactor vessel are necessary to successfully continue the history of nuclear vessel reliability in service. In addition, certain questions require further study.

Questions Requiring Additional Research

Some of the unresolved questions relative to radiation embrittlement of pressure-vessel steels, current and projected higher-alloy, higher-strength compositions, include:

1. Fundamental examination of the role of copper, phosphorus, and other residual elements in the radiation embrittlement process.

2. Development of data to permit specifications for assuring optimum radiation resistance in the total vessel: plate, weld, and weld heat affected zone.

3. Further examination of the reasons for greater sensitivity in welds than in plate.

4. Broader application and integration of recent improvements in neutron dosimetry and their extension through computer-iterative techniques of spectrum analysis to vessel embrittlement considerations.

5. Further evaluation of the yield strength versus fracture resistance (shelf drop) with very severe irradiation, that is, examination of the pattern of fracture resistance reductions and yield strength increases described above and the implications of this pattern to the total fracture resistance of the vessel.

6. Determination of the effect of radiation on fatigue crack propagation characteristics of pressure-vessel steels and weldments.

7. Establishment of an optimum balance of compositional and microstructural factors in producing, most economically, steels resistant to radiation.

8. Examination of the causes and possible corrections for observed metastability associated with elevated-temperature irradiations of certain alloy steels.

9. Fundamental evaluation of the role of thermal treatment in correcting radiation embrittlement in irradiated steels.

10. Fuller understanding of the attenuation of neutrons in a thick-walled vessel and the implications to its reliability.

11. Further correlation of data from surveillance programs and from experimental programs to validate current conclusions relative to flux rate and spectrum effects as well as for developing assurance as to the quality of sampling and environmental measurement carried out for major surveillance programs.

12. Further examination of various time-dependent factors, such as thermal and strain aging, hydrogen embrittlement, fatigue, and corrosion, and their relationship with irradiation in changing the properties of nuclear pressure vessels.

ACKNOWLEDGMENTS

Most of the work described was supported by the U.S. Atomic Energy Commission, Division of Reactor Development and Technology, and the Office of Naval Research. The support, encouragement, and advice offered by representatives of these organizations are gratefully acknowledged. The devoted efforts of the members of the Reactor Materials Branch in conducting the NRL research described are also acknowledged with sincere appreciation.

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