Heavy-Section Steel Irradiation Program

Progress Report for
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Abstract

Maintaining the integrity of the reactor pressure vessel (RPV) in a light-water-cooled nuclear power plant is crucial in preventing and controlling severe accidents which have the potential for major contamination release. The RPV is the only key safety-related component of the plant for which a duplicate or redundant backup system does not exist. It is therefore imperative to understand and be able to predict the capabilities and limitations of the integrity inherent in the RPV. In particular, it is vital to fully understand the degree of irradiation-induced degradation of the RPV's fracture resistance which occurs during service, since without that radiation damage, it is virtually impossible to postulate a realistic scenario that would result in RPV failure.

For this reason, the Heavy-Section Steel Irradiation (HSSI) Program has been established with its primary goal to provide a thorough, quantitative assessment of the effects of neutron irradiation on the material behavior and, in particular, the fracture toughness properties of typical pressure-vessel steels as they relate to light-water reactor pressure-vessel integrity. The program includes the direct continuation of irradiation studies previously conducted within the Heavy-Section Steel Technology Program augmented by enhanced examinations of the accompanying microstructural changes. Effects of specimen size; material chemistry; product form and microstructure; irradiation fluence, flux, temperature, and spectrum; and postirradiation annealing are being examined on a wide range of fracture properties. The HSSI Program is arranged into ten tasks: (1) program management, (2) K_{IC} curve shift in high-copper welds, (3) K_{Ia} curve shift in high-copper welds, (4) irradiation effects on cladding, (5) K_{IC} and K_{Ia} curve shifts in low upper-shelf (LUS) welds, (6) irradiation effects in a commercial LUS weld, (7) microstructural analysis of irradiation effects, (8) in-service aged material evaluations, (9) correlation monitor materials, and (10) special technical assistance.

During this period, a final report on the effects of neutron irradiation on the fracture toughness temperature shift and the shape of the fracture toughness (K_{IC}) curve for two submerged-arc welds (SAWs) was prepared and submitted to the Nuclear Regulatory Commission (NRC) for publication. Duplex crack-arrest specimens from Phase II of the K_{Ia} program on the same two SAWs were modified, tested, and evaluated, completing this portion of the program. A special facility for annealing irradiated Charpy V-notch (CVN) specimens was designed, manufactured, and verified to anneal existing irradiated SAW CVN specimens that were irradiated in the Fifth Irradiation Series. A data base on the irradiation, annealing, and reirradiation response of pressure-vessel steels was compiled. All unirradiated material determinations of copper variation, RT_{NDT} , and CVN upper-shelf energy throughout the LUS welds from the Midland Reactor vessel were completed. Irradiation of the first large-scale capsule, containing 25 1TC(T) and 24 1/2TC(T) Midland weld specimens, was begun in the University of Michigan Ford Nuclear Reactor and atom-probe field-ion microscopy of its precipitates, or clusters, and measure of the matrix copper content in the beltline weld completed. Initial modeling of the formation and evolution of clusters and determination of the degree to which they contribute to radiation hardening were completed and a report issued. A detailed inventory of all remaining correlation monitor material was completed. Letter reports were prepared and sent to the NRC that described various aspects of uncertainties in the embrittlement assessment of the Yankee Reactor vessel. Dosimetry measurements were performed for the High Flux Isotope Reactor location designated key 7, showing overall good correspondence with the recently calculated thermal and fast flux values but inexplicably higher fast flux values for Np and Be monitors.

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Preface

The primary goal of the Heavy-Section Steel Irradiation (HSSI) Program is to provide a thorough, quantitative assessment of the effects of neutron irradiation on the material behavior and, in particular, the fracture toughness properties of typical pressure-vessel steels as they relate to light-water reactor pressure vessel (RPV) integrity. The program includes studies of the effects of irradiation on the degradation of mechanical and fracture properties of vessel materials augmented by enhanced examinations and modeling of the accompanying microstructural changes. Effects of specimen size; material chemistry; product form and microstructure; irradiation fluence, flux, temperature, and spectrum; and postirradiation annealing are being examined on a wide range of fracture properties. Results from the HSSI studies will be incorporated into codes and standards directly applicable to resolving major regulatory issues which involve RPV irradiation embrittlement such as pressurized-thermal shock, operating pressure-temperature limits, low-temperature overpressurization, and the specialized problems associated with low upper-shelf welds.

This HSSI Program progress report covers work performed from October 1991 through September 1992. The work performed by Oak Ridge National Laboratory (ORNL) is managed by the Metals and Ceramics (M&C) Division of ORNL. Major tasks at ORNL are carried out by the M&C, Computing Applications, and Engineering Technology Divisions.

Previous HSSI Progress Reports in this series are:

NUREG/CR-5591, Vol. 1, No. 1
(ORNL/TM-11568/V1&N1)
NUREG/CR-5591, Vol. 1, No. 2
(ORNL/TM-11568/V1&N2)
NUREG/CR-5591, Vol. 2, No. 1
(ORNL/TM-11568/V2&N1)
NUREG/CR-5591, Vol. 2, No. 2
(ORNL/TM-11568/V2&N2)

Some of the series of irradiation studies conducted within the HSSI Program were begun under the Heavy-Section Steel Technology (HSST) Program prior to the separation of the two programs in 1989. Previous HSST Program progress reports contain much information on the irradiation assessments being continued by the HSSI Program as well as earlier related studies. The HSST Program progress reports issued before formation of the HSSI Program are also tabulated here as a convenience to the reader.

ORNL-4176
ORNL-4315
ORNL-4377
ORNL-4463
ORNL-4512
ORNL-4590
ORNL-4653
ORNL-4681
ORNL-4764
ORNL-4816
ORNL-4855
ORNL-4918
ORNL-4971
ORNL/TM-4655 (Vol. II)
ORNL/TM-4729 (Vol. II)
ORNL/TM-4805 (Vol. II)
ORNL/TM-4914 (Vol. II)
ORNL/TM-5021 (Vol. II)
ORNL/TM-5170
ORNL/NUREG/TM-3
ORNL/NUREG/TM-28
ORNL/NUREG/TM-49

ORNL/NUREG/TM-64
 ORNL/NUREG/TM-94
 ORNL/NUREG/TM-120
 ORNL/NUREG/TM-147
 ORNL/NUREG/TM-166
 ORNL/NUREG/TM-194
 ORNL/NUREG/TM-209
 ORNL/NUREG/TM-239
 NUREG/CR-0476 (ORNL/NUREG/TM-275)
 NUREG/CR-0656 (ORNL/NUREG/TM-298)
 NUREG/CR-0818 (ORNL/NUREG/TM-324)
 NUREG/CR-0980 (ORNL/NUREG/TM-347)
 NUREG/CR-1197 (ORNL/NUREG/TM-370)
 NUREG/CR-1305 (ORNL/NUREG/TM-380)
 NUREG/CR-1477 (ORNL/NUREG/TM-393)
 NUREG/CR-1627 (ORNL/NUREG/TM-401)
 NUREG/CR-1806 (ORNL/NUREG/TM-419)
 NUREG/CR-1941 (ORNL/NUREG/TM-437)
 NUREG/CR-2141, Vol. 1 (ORNL/TM-7822)
 NUREG/CR-2141, Vol. 2 (ORNL/TM-7955)
 NUREG/CR-2141, Vol. 3 (ORNL/TM-8145)
 NUREG/CR-2141, Vol. 4 (ORNL/TM-8252)
 NUREG/CR-2751, Vol. 1 (ORNL/TM-8369/V1)
 NUREG/CR-2751, Vol. 2 (ORNL/TM-8369/V2)
 NUREG/CR-2751, Vol. 3 (ORNL/TM-8369/V3)
 NUREG/CR-2751, Vol. 4 (ORNL/TM-8369/V4)
 NUREG/CR-3334, Vol. 1 (ORNL/TM-8787/V1)
 NUREG/CR-3334, Vol. 2 (ORNL/TM-8787/V2)
 NUREG/CR-3334, Vol. 3 (ORNL/TM-8787/V3)
 NUREG/CR-3744, Vol. 1 (ORNL/TM-9154/V1)
 NUREG/CR-3744, Vol. 2 (ORNL/TM-9154/V2)
 NUREG/CR-4219, Vol. 1 (ORNL/TM-9593/V1)
 NUREG/CR-4219, Vol. 2 (ORNL/TM-9593/V2)
 NUREG/CR-4219, Vol. 3, No. 1
 (ORNL/TM-9593/V3&N1)
 NUREG/CR-4219, Vol. 3, No. 2
 (ORNL/TM-9593/V3&N2)
 NUREG/CR-4219, Vol. 4, No. 1
 (ORNL/TM-9593/V4&N1)
 NUREG/CR-4219, Vol. 4, No. 2
 (ORNL/TM-9593/V4&N2)
 NUREG/CR-4219, Vol. 5, No. 1
 (ORNL/TM-9593/V5&N1)
 NUREG/CR-4219, Vol. 5, No. 2
 (ORNL/TM-9593/V5&N2)
 NUREG/CR-4219, Vol. 6, No. 1
 (ORNL/TM-9593/V6&N1)
 NUREG/CR-4219, Vol. 6, No. 2
 (ORNL/TM-9593/V6&N2)

Summary

1. Program Management

The Heavy-Section Steel Irradiation (HSSI) Program is arranged into ten tasks: (1) program management, (2) K_{Ic} curve shift in high-copper welds, (3) K_{Ia} curve shift in high-copper welds, (4) irradiation effects on cladding, (5) K_{Ic} and K_{Ia} curve shifts in low upper-shelf (LUS) welds, (6) irradiation effects in a commercial LUS weld, (7) microstructural analysis of irradiation effects, (8) in-service aged material evaluations, (9) correlation monitor materials, and (10) special technical assistance. Report chapters correspond to the tasks. The work is performed by the Oak Ridge National Laboratory (ORNL). During the report period, 16 technical presentations were given, 6 technical papers were published, and 5 letter reports were issued.

2. K_{Ic} Curve Shift in High-Copper Welds

The two primary objectives of this study were to determine the effects of neutron irradiation on the fracture toughness temperature shift and the shape of the fracture toughness (K_{Ic}) curve for two submerged-arc welds (SAWs) with copper contents of 0.23 and 0.31%. All the originally planned experimental and analytical tasks for this project have been completed. A final report has been prepared and submitted to the Nuclear Regulatory Commission (NRC) for publication. Analyses revealed that the fracture toughness shifts (at 100 MPa $\sqrt{m}$) exceeded the Charpy shifts for both welds. Analyses of curve shape changes indicate slight irradiation-induced decreases in the slopes of the fracture toughness curves, especially for the higher copper weld.

3. K_{Ia} Curve Shift in High-Copper Welds

The objectives of the Sixth Irradiation Series are to determine the K_{Ia} curve shifts and shapes for two high-copper, SAWs. The program is being conducted in two phases. In Phase I, 36 weld-embrittled crack-arrest specimens were tested, and detailed results with some preliminary conclusions have been published and a summary presented in a previous semiannual progress report. Phase II of the K_{Ia} program has been completed; duplex crack-arrest specimens have been modified and tested and the results presented. A specially adapted, remotely operable lathe was placed in the hot cell and used to modify the irradiated duplex specimens. A brief update is also given about activities involving testing of irradiated specimens for The Italian Committee for Research and Development of Nuclear Energy and Alternative Energies (ENEA).

4. Irradiation Effects in Cladding

The objective of this series is to obtain toughness properties of stainless steel cladding in the unirradiated and irradiated conditions. The properties obtained include tensile, Charpy V-notch (CVN) impact, and J-integral toughness. The goal is to evaluate the fracture resistance of irradiated weld-metal cladding representative of that used in early pressurized-water reactors (PWRs). The fracture properties are needed for detailed integrity analyses of vessels during overcooling situations. There was no significant activity within this task during this reporting period. A limited number of precracked CVN specimens of three-wire cladding will be irradiated as part of the HSSI Tenth Irradiation Series and will be tested in 1994.

5. K_{Ic} and K_{Ia} Curve Shift and Annealing in LUS Welds

Two irradiation series will be performed within this task. The primary objective of Series 8 is to examine the K_{Ic} and K_{Ia} for LUS high-copper weld metal irradiated at 288°C (550°F), with particular emphasis on the shift and change in shape of the American Society of Mechanical Engineers curves following irradiation. The purpose of the Ninth Irradiation Series is to evaluate the correlation between fracture toughness and CVN impact energy during irradiation, annealing, and reirradiation. During this reporting period, a special container for annealing irradiated CVN specimens has been designed, manufactured, and verified. It will be used in an available

controlled-atmosphere furnace to anneal at 454°C, for 168 h, some of the existing inventory of undersize HSSI weld 73W CVN specimens that were irradiated in the Fifth Irradiation Series capsules. A data base on the irradiation, annealing, and reirradiation response of pressure-vessel steels has been compiled. It was used to evaluate the annealing response of pressure-vessel steels irradiated at 260°C. It was also transmitted at the request of the NRC to both the NRC and to Modeling and Computing Associates.

6. Irradiation Effects in a Commercial LUS Weld

The primary objective of the Tenth Irradiation Series is to investigate the postirradiation fracture toughness of the SAW from the Midland Unit 1 reactor vessel. The weld from that vessel is of considerable interest because it carries the Babcock and Wilcox Co. designation WF-70, an SAW fabricated with a specific heat of weld wire (heat 72105) and specific lot of welding flux S (lot 8669). The WF-70 weld was fabricated using copper-coated wire and Linde 80 flux and is known to be LUS, high-copper weld. Major objectives of the unirradiated material characterization include determinations of copper variation, RT_{NDT} , and CVN upper-shelf energy throughout the welds (25 individual CVN curves were obtained). This part of the unirradiated characterization testing has been completed and recently reported. Major observations and conclusions are given in this progress report and show, for example, very wide ranges in RT_{NDT} (from -20 to 37°C) and copper content (0.21 to 0.46%) throughout the welds. Regarding the fracture toughness testing and material irradiations, all specimens have been fabricated, and the first large-scale capsule, containing 25 1TC(T) and 24 1/2TC(T) specimens, is currently under irradiation in the University of Michigan Ford Nuclear Reactor.

7. Microstructural Analysis and Modeling

The objective of this task is to provide an enhanced capability to interpolate and extrapolate the degree of radiation embrittlement beyond existing data bases. To accomplish this, a combination of studies examining ultrafine-scale radiation-induced damage and developing models based on fundamental mechanisms are being performed. During this reporting period, the beltline weld of the Midland Unit 1 reactor pressure vessel was examined by atom-probe field-ion microscopy to characterize the nature of its precipitates, or clusters, and measure the matrix copper content. The modeling work initiated under this task to examine the formation and evolution of clusters and determine the degree to which they contribute to radiation hardening reached interim conclusions, and a report describing those conclusions was issued. A new collaboration was initiated between ORNL staff and researchers at the Harwell Laboratory and the University of Liverpool in the United Kingdom to investigate high-energy cascades in iron, using the MOLDY molecular dynamics simulation (or MDS) code, and a newly developed interatomic potential for iron, and experiments to examine effects of flux and spectrum at low temperature began in earnest.

8. In-Service Aged Material Evaluations

The overall objective of this task is to assess the service-induced degradation of fracture resistance through examination of components exposed during in-nuclear-plant operation. The initial focus of this task is to augment the existing hot-cell testing capability available to the HSSI Program with remote machining capabilities for the fabrication of specimens from samples of activated steel obtained from service-exposed components. During this reporting period, a decision was made to pursue the acquisition of a computer numerically controlled machining center suitable for hot cell operations, and bids for suitable machines were let.

9. Correlation Monitor Materials

This is a new task that has been established with the explicit purpose of ensuring the continued availability of the pedigreed and extremely well-characterized material now required for inclusion in all additional and future surveillance capsules in commercial light-water reactors. Having recognized that the only remaining materials qualified for use as a correlation monitor in reactor surveillance capsules are the pieces remaining from the early Heavy-Section Steel Technology (HSST) plates 01, 02, and 03, this task will provide for cataloging, archiving, and distributing the material on behalf of the NRC. During this reporting period, a detailed inventory of all remaining correlation monitor material was completed, and a piece of the correlation monitor material from plate HSST 02 was shipped to the University of Missouri for research on subsized CVN specimens.

10. Special Technical Assistance

This task was established during the current reporting period to explicitly emphasize and provide performance and financial monitoring of various analytical and experimental investigations conducted to support the NRC in resolving short-term, high-priority regulatory and research issues. The current activities being performed as part of this task include providing expert guidance to the NRC staff regarding irradiation effects issues surrounding the embrittlement of the Yankee Reactor pressure vessel and providing dosimetry measurements for various surveillance specimen locations within the vessel of the High Flux Isotope Reactor (HFIR) at ORNL. With regard to the evaluation of embrittlement of the Yankee Reactor vessel, three letter reports were prepared and sent to the NRC that described uncertainties in the assessment of the vessel, analyses of the power reactor embrittlement base, chemistry and orientation effects, and statistical considerations in vessel sampling. Dosimetry measurements were performed for the HFIR location designated key 7. Experimental data showed overall good correspondence with the recently calculated thermal and fast flux values, but measurements for Np and Be monitors gave inexplicably higher fast flux values.

Heavy-Section Steel Irradiation Program Progress Report for October 1991 through September 1992*,†

W. R. Corwin

1. Program Management

The Heavy-Section Steel Irradiation (HSSI) Program, a major safety program sponsored by the Nuclear Regulatory Commission (NRC) at Oak Ridge National Laboratory (ORNL), is an engineering research activity devoted to providing a thorough, quantitative assessment of the effects of neutron irradiation on the material behavior, particularly the fracture toughness properties, of typical pressure-vessel steels as they relate to light-water reactor (LWR) pressure-vessel integrity. The program centers on experimental assessments of irradiation-induced embrittlement [including the completion of certain irradiation studies previously conducted by the Heavy-Section Steel Technology (HSST) Program] augmented by detailed examinations and modeling of the accompanying microstructural changes. Effects of specimen size; material chemistry; product form and microstructure; irradiation fluence, flux, temperature, and spectrum; and postirradiation annealing are being examined on a wide range of fracture properties. Fracture toughness (K_{IC} and J_{IC}), crack-arrest toughness (K_{Ia}), ductile tearing resistance (dJ/da), Charpy V-notch (CVN) impact energy, drop-weight nil-ductility transition (NDT), and tensile properties are included. Models based on observations of radiation-induced microstructural changes using the field-ion microprobe and the high-resolution transmission electron microscope are being developed to provide a firm basis for extrapolating the measured changes in fracture properties to wide ranges of irradiation conditions. The principal materials examined within the HSSI Program are high-copper welds because their postirradiation properties frequently limit the continued safe operation of commercial reactor pressure vessels (RPVs). In addition, a limited effort will focus on stainless steel weld-overlay cladding typical of that used on the inner surfaces of RPVs because its postirradiation fracture properties have the potential for strongly affecting the extension of small surface flaws during overcooling transients.

Results from the HSSI studies will be integrated to aid in resolving major regulatory issues facing the NRC. Those issues involve RPV irradiation embrittlement such as pressurized-thermal shock, operating pressure-temperature limits, low-temperature overpressurization, and the specialized problems associated with low upper-shelf (LUS) welds. Together, the results of these studies also provide guidance and bases for evaluating the overall aging behavior of LWR pressure vessels.

The program is coordinated with those of other government agencies and the manufacturing and utility sectors of the nuclear power industry in the United States and abroad. The overall objective is the quantification of irradiation effects for safety assessments of regulatory agencies, professional code-writing bodies, and the nuclear power industry.

The program is broken down into one task responsible for overall program management and nine technical tasks: (1) program management, (2) K_{IC} curve shift in high-copper welds, (3) K_{Ia} curve shift in high-copper welds, (4) irradiation effects on cladding, (5) K_{IC} and K_{Ia} curve shift in LUS welds, (6) irradiation effects in a commercial LUS weld, (7) microstructural analysis of irradiation effects, (8) in-service aged material evaluations, (9) correlation monitor materials, and (10) special technical assistance. Accordingly, the chapters of this progress report correspond to these ten tasks.

During this period, 13 program briefings, reviews, or presentations were made by the HSSI staff during program reviews and visits with NRC staff or others. Six technical papers¹⁻⁶ were published, and five letter reports⁷⁻¹¹ issued. In addition, 16 technical presentations were made.¹²⁻²⁸

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2. K_{Ic} Curve Shift in High-Copper Welds

R. K. Nanstad, D. E. McCabe, and F. M. Haggag

All the originally planned experimental and analytical tasks for this project have been completed. A final report has been prepared and submitted to NRC for publication.¹ The section entitled "Summary, Discussion, and Conclusions" is reproduced below:

Summary, Discussion, and Conclusions

The two primary objectives of this study were to determine the effects of neutron irradiation on the fracture-toughness temperature shift and the shape of the fracture toughness (K_{Ic}) curve for two SAWs with copper contents of 0.23 and 0.31%. The fracture toughness values from small cleavage pop-ins suggest that the pop-ins observed in this study are significant in that they indicate propensity for cleavage fracture in the test specimen. The subject of cleavage pop-ins in laboratory test specimens and their relevance to reactor vessel integrity is a major technical issue in itself. It is dealt with in standard testing procedures to the degree that arbitrary definitions of significance based on pop-in size have been proposed. Although those definitions may be found to be adequate, they have not been rigorously founded. The observations in this study give strong indications that the initial cleavage pop-ins that occurred could not be considered outliers from an engineering viewpoint and certainly not from a statistical viewpoint. At, or near, test temperatures with low toughness pop-ins, there were often cleavage failures. Thus, in the absence of technical justifications, the initial cleavage pop-ins observed in this study were considered significant to the structural integrity of the test specimen. To some degree, the quantitative results from this study are dependent on the inclusion of those pop-ins. However, exclusion of those results from the data base would not remove the observed differences between the fracture toughness and Charpy shifts. Resolution of this issue is beyond the scope of this report.

Regarding the irradiation-induced temperature shift, statistical analyses and curve fitting using nonlinear regressions showed that the temperature shifts at a fracture toughness of 100 MPa $\sqrt{m}$ were greater than those at a Charpy energy of 41 J for both welds. The shifts and the differences between the Charpy and fracture toughness shifts were greater for the higher copper weld. The amounts of the 100-MPa $\sqrt{m}$ shifts were dependent on the particular model used; the three- and two-parameter nonlinear fits gave shifts of about 94 and 100°C, while the linearized two-parameter (with fixed intercept) fit gave shifts of about 83 and 99°C for 72W and 73W, respectively. Thus, for weld 72W, the 100-MPa $\sqrt{m}$ shift is from 11 to 22°C greater than the Charpy 41-J shift, while it is about 18°C greater for weld 73W. Analyses of the data with elimination of those results from specimens that experienced significant precleavage stable ductile tearing did not change the overall observations regarding the shifts of the fracture toughness curves. The same observation applies to analyses of β_{Ic} -adjusted data and data satisfying the Anderson and Dodds² definition for the limit of small-scale yielding (thickness > 200 x J/s_y).

Determination of the standard errors associated with the irradiation-induced Charpy impact and fracture-toughness mean shifts revealed standard errors ranging from ± 3 to $\pm 5^\circ\text{C}$, reflecting the relatively large data sets available for analysis. Furthermore, using the linearized model, the temperature intervals associated with one standard deviation on the predicted value of fracture toughness, 37 and 30°C, are greater than those for the Charpy impact data, 20 and 22° for 72W and 73W, respectively. Using the linear fits, the 100-MPa $\sqrt{m}$ temperature shifts, defined as the mean shift plus one standard deviation on the data, are 120 and 129°C; these values can be compared with similarly defined Charpy 41-J temperature shifts of 92 and 102°C, for 72W and 73W, respectively. The Charpy 68-J transition temperature shifts are in much better agreement with the 100-MPa $\sqrt{m}$ fracture toughness shifts than are the 41-J shifts, but this observation is presently regarded as fortuitous in the absence of other similar experimental evidence.

In a review of irradiation-induced Charpy and fracture-toughness shifts, Hiser³ also used a mean curve-fitting technique and compared the reported results from plates, forgings, and welds. The analysis revealed that for fracture-toughness shifts relative to those for Charpy impact, plates were about 15°C greater, forgings were about 24°C greater, and welds were about the same. The overall average comparison was that the 100-MPa $\sqrt{m}$ fracture toughness shifts were about 10°C greater than the Charpy 41-J shifts. The report also showed that one standard deviation temperature intervals between the means and lower-boundary curves ranged from about 11 to 64°C, with an overall value of about 32°C, similar to those observed in this study. As noted in the report, however, many of the fracture-toughness curves were developed with six or even fewer data; thus, the uncertainties in those

of the fracture-toughness curves were developed with six or even fewer data; thus, the uncertainties in those cases would be quite high. However, the overall observations concerning fracture-toughness shifts are generally similar to those of this study.

To reiterate a previous point, a comparison between the irradiation-induced shifts for welds 72W and 73W and those predicted by Regulatory Guide (RG) 1.99 (Rev. 2) is interesting but not relevant to the objectives of this study. Even though the regulatory guide may overpredict the CVN shifts for these welds, there are many materials for which RG 1.99 (Rev. 2) underpredicts the CVN shifts. Because the regulatory guide is based solely on CVN data, and given the observation that the K_{IC} shift may be greater than the CVN shift, a regulatory guide analysis may underpredict the fracture-toughness shift for many materials.

Regarding the shape of the fracture-toughness curve, the results from curve-fitting data are somewhat mixed. Because the construction of a lower-bound curve to fracture-toughness data can be highly subjective, nonlinear regression analyses were performed to obtain mean fits to the data. The two-parameter (with fixed intercept) and two-parameter nonlinear mean fits did not indicate degradation of shape, but those models are not amenable to systematic quantitative evaluations of curve slope changes. Thus, a linearized two-parameter model with fixed intercept was used and did indicate some decreases in the slopes, with the higher copper weld 73W exhibiting a greater change than for the lower copper weld 72W. The change in slope for weld 73W, in fact, was more than half that needed to achieve the same slope as the American Society for Mechanical Engineers (ASME) K_{Ia} curve. Using a 95% confidence criterion, however, the statistical significance of those changes was mixed. Thus, although the shape changes are not dramatic, the analyses do indicate that degradation in the slopes of the mean fracture-toughness curves occurred as a consequence of irradiation. The implication of that observation is that a bounding curve would, therefore, also change.

The ASME K_{IC} curve, however, is a lower-bound curve to the data used for its construction and presumably was not developed from evaluation of a mean curve. As mentioned, the construction of bounding curves can be highly subjective. Curves manually constructed to lower bound all the data do indicate a slope decrease, especially for weld 73W, but the construction of such a curve is very subjective beyond about 100-MPa $\sqrt{m}$. In the previous section, various methods were used to develop bounding curves, e.g., 5 percentile or 95% confidence curves. Figures 2.1(a) and (b) show plots of the irradiated fracture-toughness data and various curves for 72W and 73W, respectively. The ASME K_{IC} curve is shown for the unirradiated condition and for the irradiated condition after shifting the curve upward in temperature equal to the Charpy 41-J shift (DTT_{41}). The $K_{0.05}$ curve from the Wallin procedure, using a fixed K_{min} value of 20 MPa $\sqrt{m}$ ($K_{0.05-20}$), bounds all the data in both cases and has a substantially lower slope than the K_{IC} curve and the $K_{0.05}$ curve developed with a variable K_{min} ($K_{0.05-vary}$). As would be expected, some K_{JC} results fall below the $K_{0.05-vary}$ curve. The $K_{0.05-vary}$ curves are substantially above the $K_{0.05-20}$ curves. The other curve shown in each plot is the K_{IC} curve shifted upward in temperature until it just bounds all the data. The shifts necessary to bound the data in each case are about the same as reported earlier for the difference between the Charpy 41-J and K_{JC} 100-MPa $\sqrt{m}$ shifts, meaning that the shifts of the mean K_{JC} curves are about the same as the shifts of the bounding curves. The large difference in the slopes of the K_{IC} and $K_{0.05-20}$ curves is apparent in those plots. Figure 2.2 shows a similar plot of the combined 72W and 73W irradiated data vs $T - RT_{NDT}$.

To enable an easier comparison of the shape of the various curves, Figure 2.3 shows the same data and curves as in Figure 1(b) for weld 73W, but each curve has been shifted until it just bounds all the data. Both of the $K_{0.05}$ curves have considerably lower slopes than the ASME K_{IC} curve with the $K_{0.05-20}$ curve having the lowest slope as stated earlier. Relative to the actual data, the $K_{0.05}$ curves are better descriptors of the lower bound up to about 100 MPa $\sqrt{m}$. Above that level, however, those curves appear to be overly conservative relative to the available data. The ASME K_{Ia} , crack-arrest toughness, curve has a somewhat lower slope than the K_{IC} curve and is plotted in Figure 2.4 for comparison with the curves described in Figure 2.3. Again, the K_{Ia} curve was positioned until it just bounds all the data. As seen in the figure, the K_{Ia} curve is higher than both the $K_{0.05}$ curves throughout the transition region. Its slope, in fact, is closer to that data of the K_{IC} curve than to the $K_{0.05-vary}$ curve.

Kaun and Koehring⁴ irradiated 4T specimens of a reactor vessel steel weld metal at various fluences up to about 3×10^{19} neutrons/cm² (<1 MeV) and stated that increasing neutron irradiation not only shifted the K_{IC} curves, but also reduced the slopes. They did not provide a quantitative evaluation of the changes except for a plot of the fitted curves. The material experienced a 100-MPa $\sqrt{m}$ fracture-toughness shift of about 175°C (315°F) but a 200-MPa $\sqrt{m}$ shift of about 205°C (369°F). Thus, a substantial degradation in the shape of the curve occurred as a result of irradiation. That material experienced a substantially greater shift than those for 72W and 73W, and the

Figure 2.1.

Fracture toughness, K_{Jc} , versus test temperature for irradiated HSSI welds: (a) 72W and (b) 73W comparing the ASME K_{Ic} curve shifted by the Charpy impact 41-J shift with Weibull-based 5 percentile curves. The $K_{0.05}$ curves were developed using a Weibull method wherein the K_{min} parameter is either fixed at 20 MPa·√m ($K_{min} = 20$) or varied (K_{min} Vary) by setting it equal to $K_{Jc\text{low}} - 5$ MPa·√m, where $K_{Jc\text{low}}$ is the lowest result at each temperature analyzed. The K_{Ic} lower-bound curve is simply the K_{Ic} curve shifted upward in temperature until it bounds all the data.

Figure 2.2

Fracture toughness, K_{IC} , versus $T - RT_{NDT}$ for combined irradiated HSSI welds 72W and 73W comparing the ASME K_{IC} curve shifted by the Charpy impact 41-J shift with Weibull-based 5 percentile curves. The $K_{0.05}$ curves were developed using a Weibull method wherein the K_{min} parameter is either fixed at 20 MPa·√m ($K_{min} = 20$) or varied (K_{min} Vary) by setting it equal to $K_{Jclow} - 5$ MPa·√m, where K_{Jclow} is the lowest result at each temperature analyzed. The K_{IC} lower-bound curves simply the K_{IC} curve shifted upward in temperature until it bounds all the data.

Figure 2.3.

Fracture toughness, K_{IC} , versus test temperature for irradiated HSSI weld 73W comparing the ASME K_{IC} curve shifted by the Charpy impact 41-J shift with Weibull-based 5 percentile curves. The $K_{0.05}$ curves were developed using a Weibull method wherein the K_{min} parameter is either fixed at 20 MPa·√m ($K_{min} = 20$) or varied (K_{min} Vary) by setting it equal to $K_{Jclow} - 5$ MPa·√m, where K_{Jclow} is the lowest result at each temperature analyzed. The K_{IC} lower-bound curve is simply the K_{IC} curve shifted upward in temperature until it bounds all the data and is compared with the $K_{0.05}$ curves similarly shifted.

Figure 2.4.

Fracture toughness, K_{Jc} , versus test temperature for irradiated HSSI weld 73W comparing the ASME K_{Ic} curve shifted by the Charpy impact 41-J shift with Weibull-based 5 percentile curves and the ASME K_{Ia} curve. The $K_{0.05}$ curves were developed using a Weibull method wherein the K_{min} parameter is either fixed at 20 MPa·√m ($K_{min} = 20$) or varied (K_{min} Vary) by setting it equal to $K_{Jc\text{low}} - 5$ MPa·√m, where $K_{Jc\text{low}}$ is the lowest result at each temperature analyzed. The K_{Ic} lower-bound curves are simply the K_{Ic} and K_{Ia} curves shifted upward in temperature until they bound all the data and are compared with the $K_{0.05}$ curves similarly shifted.

apparent change in the fracture-toughness curve was also greater. This observation, of course, gives rise to the concern that degradation of the K_{Ic} curve shape increases with increasing embrittlement.

Of course, concerns about curve shape changes can be accounted for by applying large enough shifts to the K_{Ic} curve, but such a practice begs the issue of how good accuracy can be developed when the trend line (curve shape) does not fit the real fracture-toughness trend for the irradiated condition. The data in the present case indicate that the K_{Ic} curve shifted by the 41-J CVN plus the one standard deviation "margin" of RG 1.99 (Rev. 2) did not provide a lower bound to the data. This is consistent with the observations of Hiser³ noted earlier. Thus, more margin adjustment would be needed. The difficulty is that added margin to cover high-toughness K_{Jc} values will result in overconservatism in the lower transition region. Therefore, shallower curves such as the five percentile curves based on Weibull analyses or the K_{Ia} curve deserve consideration. A reasonable argument can be made for use of the K_{Ia} curve shape on the basis that irradiation damage tends to increase material strength and consequently reduce the strain rate sensitivity of RPV steels such that K_{Ic} values tend toward agreement with K_{Ia} values. Preliminary observations from the HSSI Sixth Irradiation Series on crack-arrest toughness indicate no irradiation-induced curve shape changes in the K_{Ia} curve.⁵ One consideration, then, would be the use of the K_{Ia} curve shape to describe the irradiated K_{Ic} curve for materials to exhibit irradiation-induced toughness shifts above some prescribed amount.

Regarding specimen size effects, the exponential curve fits to the unirradiated data separated by specimen size (1T, 2T, and 4T) indicated a size effect on the fracture toughness results with smaller specimens showing higher average toughness. It is also true, however, that the smaller specimens often exhibit the lower toughness values

at a given temperature. An important result was shown earlier in that there was no apparent size effect on the resistance curve of fracture toughness versus precleavage ductile tearing. In that regard, Figure 2.5 shows the " β_{Ic} -adjusted" data for irradiated weld 73W with the K_{Ic} curve shifted to just bound the data. At 100 MPa $\cdot\sqrt{m}$, the temperature difference between that curve and the K_{Ic} curve shifted by the Charpy 41-J shift is about 26°C, compared with a shift of about 18°C required to similarly bound the unadjusted data. Specimen size effects in the transition region, such as those observed here, is a subject receiving considerable attention worldwide. Such results will receive continuing attention, especially regarding the use of small fracture-toughness specimens from surveillance capsules.

Figure 2.5. Fracture toughness, K_{Bc} (β_{Ic} -adjusted), versus test temperature for irradiated HSSI weld 73W comparing the ASME K_{Ic} curve shifted by the Charpy impact 41-J shift and shifted until it bounds all the data.

The principal conclusions drawn from this study are as follows:

1. Cleavage pop-ins were significant in that they indicate lack of resistance to cleavage fracture in the test specimen at the stress-intensity level of the pop-in and under the particular test conditions.
2. The fracture-toughness temperature shifts at 100 MPa $\cdot\sqrt{m}$ of about 83 and 100°C (149 and 180°F) are about 11 and 18°C (20 and 32°F) greater than the corresponding Charpy 41-J shifts but are similar to the Charpy 68-J shifts.
3. The temperature intervals associated with one standard deviation on the fracture-toughness results are greater than the corresponding intervals for the Charpy results. Therefore, the mean 100 MPa $\cdot\sqrt{m}$ shifts plus one standard deviation temperature interval are substantially greater than the corresponding shifts for the Charpy results.

4. The measured Charpy 41-J shifts plus the margin (one standard deviation) in RG 1.99 (Rev. 2) do not bound all the fracture-toughness data.
5. Mean curve fits to the fracture-toughness data with a linearized two-parameter exponential (with fixed intercept) indicate irradiation-induced curve shape changes for both welds with the change being greater for weld 73W (0.31% Cu) than that for weld 72W (0.23% Cu).
6. Weibull-based analyses provide reasonable lower-bound curves but can be overly conservative at the higher toughness levels; a method incorporating a variable K_{min} parameter in the three-parameter Weibull model offers an alternative method for development of acceptable lower-bound curves.
7. The ASME K_{Ia} curve shape should be considered as a potential curve shape for highly radiation-sensitive materials that undergo large shifts.
8. Although statistically significant specimen size effects were observed, analyses of results with precleavage stable ductile tearing indicated that the observed specimen size effects were due to constraint and not effects of ductile tearing.
9. Limiting the results to those which did not experience significant stable ductile tearing, to those satisfying the definition of small-scale yielding, and β_{IC} -adjusted data did not change the results of the analyses regarding shifts and shape changes of the fracture-toughness curves.

An opportunity is being taken to irradiate 1/2TC(T) specimens of the above materials in the capsule for Task 6. This will be an opportunity to test recent developments in statistical methods designed to establish transition temperatures and lower-bound toughness from surveillance capsule specimens. There appears to be compelling evidence that the lower-bound toughness can be determined without the aid of large test specimens. Currently, lower-bound trends before and after irradiation at 95% confidence have been established with 1TC(T) specimen data (see Figures 2.6 and 2.7).

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Figure 2.6. Fifth Irradiation Series K_{Jc} data (unirradiated, weld 72W) fit to a universal median curve.

Figure 2.7. Fifth Irradiation Series K_{Jc} data (irradiated, welds 72W and 73W) fit to a universal median curve.

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†Available in public technical libraries.

3. K_{Ia} Curve Shift in High-Copper Welds (Phase II)

S. K. Iskander, R. K. Nanstad, and E. T. Manneschildt

3.1 Introduction

The objective of the Sixth Irradiation Series is to determine the effect of neutron irradiation on the shift and shape of the K_{Ia} versus (T - RT_{NDT}) curve, where K_{Ia} is the plane-strain crack-arrest fracture toughness, T is the temperature, and RT_{NDT} is the reference NDT temperature. The Sixth Series investigates the effects of irradiation on the fracture toughness of welds, since some pressure vessels in operation have welds with copper contents and end-of-life fluences that make them susceptible to severe degradation in toughness. The amount of experimental data on the effects of irradiation on crack-arrest fracture toughness is still rather meager.¹⁻³

Two SAWs with copper contents of 0.23 and 0.31 wt %, designated HSSI welds 72W and 73W, respectively, were commercially fabricated in 220-mm-thick plate for use in both the Fifth and Sixth Irradiation Series. In the Fifth Irradiation Series, unirradiated and irradiated CVN, impact, tensile, drop-weight, and compact specimens were tested, and the results are given in refs. 4 through 6. The crack-arrest specimens for the Sixth Irradiation Series were also fabricated from the 72W and 73W welds.

A previous detailed report³ and summary paper⁷ presented the test results of the unirradiated specimens as well as 36 weld-embrittled crack-arrest specimens in Phase I of the program. In Phase II, the duplex-type crack-arrest specimens are to be tested. The crack-arrest specimens were irradiated at a nominal temperature of 288°C to a nominal fluence of 1.9×10^{19} neutrons/cm² (>1 MeV). Complete details of the dosimetric calculations are given in ref. 8. Testing is performed according to the American Society for Testing and Materials (ASTM), "Test for Determining Plane-Strain Crack-Arrest Fracture Toughness, K_{Ia}, of Ferritic Steels" (E 1221-88).

For reasons given below, the irradiated duplex crack-arrest specimens had to be modified, and a lathe was adapted to perform this modification. The 20 remaining irradiated duplex crack-arrest specimens were tested, and the values of the crack-arrest toughness obtained are given. A detailed report on Phase II has been published.⁹

3.2 Modification of the Irradiated Duplex Crack-Arrest Specimens

At the conclusion of the Phase I tests on weld-embrittled-type crack-arrest specimens, four duplex-type crack-arrest specimens were tested. All four tests were unsuccessful because crack propagation was arrested in the fusion zone between the hard 4340 steel crack-starter section and the weld metal test section. The most likely cause of the arrest is the lack of fusion between the two sections. The gap between the two regions acts like an arrester hole that is sometimes introduced in structures to stop a growing crack. It should be noted that the heat-affected zone (HAZ) is generally tougher than the surrounding material, and thus it is not uncommon for it to arrest a running flaw in duplex type specimens. The test section is electron-beam (EB) welded to the 4340 steel crack-starter section from one side, then turned to complete the weld from the other side. The specimens are 33 mm thick, and EB equipment has enough power to weld up to 100-mm-thick sections. The welding is performed from both sides to minimize the heat input and to reduce the width of the HAZ. In the case of the subject irradiated duplex crack-arrest specimens, unexpected changes in the welding procedure produced EB welds that did not penetrate to the desired 60% of the specimen thickness obtained in previous specimen fabrication. Prior to irradiation, all the duplex crack-arrest specimens were examined using X-ray radiography. The examination did not reveal the presence of any discontinuities.

From the tests of the first four specimens, it seemed highly probable that this lack of fusion existed in all the remaining specimens. In retrospect, this was indeed the case. In order to utilize these specimens, various options were considered. One was to store the specimens until such time that the irradiated weld metal could be used for some other task, as yet unknown. It was considered worthwhile to try to obtain data from these specimens, even at the risk of a large percentage of unsuccessful tests, always a possibility in crack-arrest testing. Another option was to reweld the specimens, but this did not appear feasible because of the difficulty of locating EB equipment capable of handling irradiated specimens. Even if such equipment was located, the presence of the side grooves

would make it difficult to focus the EB to the precise location of the interface. Previous experience of rewelding unirradiated specimens shows that this may be successful before the side grooves are machined.

The only option that seemed feasible was to increase the crack-starter hole diameter. The theory behind this modification is that a sufficiently large crack-driving force may cause the propagating flaw to "jump" through the sound material on either side of the unfused zone. Trial tests were made to verify this option using unirradiated specimens with an intentionally unfused zone. These tests indicated that if the unfused region is no larger than about one-third of the net section at the root of the side grooves (which is indeed the case in some of the four irradiated 72W and 73W specimens already tested and mentioned above), increasing the crack-starting hole diameter to 16 or 19 mm would increase the probability of obtaining useful data from the 20 irradiated crack-arrest specimens.

Based on this limited success, a method had to be developed to increase the diameter of an existing hole with an adjoining slot. The problem of modifying this geometry in a hardened irradiated steel had to be addressed. The new hole must be tangential to the old one to preserve the initial crack length-to-width ratio. As is well known, it is not possible to use a drill, since the existing hole would have the tendency to force the center line of the new hole to be concentric with the old one. The problem was solved by using a carbide tip end mill hole-trepanning cutter in a lathe that was specifically "configured" to accomplish this modification in the hot cells.

3.3 Results of Testing the Modified Irradiated Duplex Crack-Arrest Specimens

Twelve each of HSSI welds 72W and 73W duplex crack-arrest specimens were irradiated in HSSI Capsules 6-1 and 6-2, respectively. As mentioned above, four specimens were tested before the unfused EB weld region problem was discovered. Because all four specimens showed lack of fusion, it was considered to be a likely condition in all the specimens. The remaining 20 were eventually modified and tested, and a summary of the test results is given below.

Of the ten specimens from each of welds 72W and 73W, there were four successful tests, all with specimens from weld 72W.* The crack-arrest toughness values, K_{Ia} , the irradiation exposures, and the validity criteria, according to the ASTM "Test for Determining Plane-Strain Crack-Arrest Fracture Toughness, K_{Ia} , of Ferritic Steels" (E 1221-88), are given in Table 3.1. The K_a values for the duplex crack-arrest specimens have been plotted, together with those of the weld-embrittled specimens previously obtained,³ against the test temperature in Figure 3.1. It may be seen that the K_a values of the duplex crack-arrest specimens all fall near the upper end of the scatter specimens. It should be noted, however, that the average fluence of these four duplex crack-arrest specimens, 1.56×10^{19} neutrons/cm² (> 1 MeV), is somewhat lower than that of the weld-embrittled crack-arrest specimens, 1.88×10^{19} neutrons/cm² (> 1 MeV). Thus, the toughness values obtained from these specimens being somewhat higher than those of the weld-embrittled specimens seems reasonable.

The figure also shows two curves based on the ASME K_{Ia} equation,[†] which in SI units is

$$K_a = 29.4 + 1.344 \exp [0.0261(T - T_0 + 89)] , \quad (\text{Eq. 3-1})$$

where K_a is the crack-arrest toughness in MPa $\sqrt{m}$; T is the temperature in °C; and T_0 is a parameter, in °C. The crack-arrest toughness data were fit to the above equation with T_0 as the unknown parameter. The process was performed once with the 18 weld-embrittled crack-arrest toughness values obtained previously,³ then a second

*There was a single successful crack initiation-and-run event in a specimen from weld 73W. Unfortunately, there is sufficient reason to question the accuracy of temperature indicated, and the result has not been used.

†In the 1992 Addenda (issued Dec. 31, 1992) of the ASME Boiler and Pressure Vessel Code, the following equation (converted to SI units) for K_{Ia} is given in Article A-4000 of Section XI: $K_{Ia} = 29.4 + 13.675 \exp[0.0261(T - RT_{NDT})]$. The equation appears to be the amplification of the one given in WRC Bulletin 175 (August 1992) and not that of the K_{Ic} equation given in Article G-2000 of Section III.

Table 3.1. Irradiated Crack-Arrest toughness data for four duplex specimens from weld 72W
[average fluence and irradiation temperature were
 1.56×10^{19} neutrons/cm² (> 1 MeV) and 288°C]

Specimen	Test temperature (°C)	K _a * (MPa√m)	Irradiation temperature (°C)	Exposure values			Validity†
				Fluence (neutrons/cm ²)		Displacements per atom	
				> 1 MeV	> 0.1 MeV		
A72W50	50	82.2	288	1.53E19	1.020E20	0.0379	C, D
A72W52	75	133.0	289	1.67E19	1.150E20	0.419	
A72W61	91	137.4	287	1.56E19	1.040E20	0.385	
A72W72	75	152.8	287	1.49E19	0.978E20	0.363	

* K_a = value of stress-intensity factor shortly after arrest.

† One or more letters indicate that test results did not meet one of the minimum lengths of ASTM E 1221-88 validity criteria. Letters correspond to those in Table 2 of ASTM E 1221-88; in particular, C = specimen too thin—the standard prescribes (based on plastic zone size considerations) 37 mm (1.46 in.), and the actual thickness was 33 mm (1.3 in.); and D = insufficient crack-jump length—the standard prescribes (based on the crack-starter hole diameter) 38 mm (1.5 in.), and the actual jump was 21 mm (1.23 in.)

Figure 3.1 Crack-arrest toughness, K_a , for irradiated weld 72W showing the results of both weld-embrittled and duplex-type specimens.

time with the results of both the 18 weld-embrittled and 4 duplex crack-arrest specimens. The T_0 values were 14.2 and 11.7°C for the weld-embrittled and both specimen types, respectively. The lower T_0 value reflects the influence of the higher K_a values of the duplex crack-arrest specimens compared to those of the weld-embrittled specimens.

The experimentally obtained crack-arrest toughness values for both unirradiated and irradiated 72W weld metal and for weld-embrittled and duplex-type specimens are plotted in Figure 3.2. Also shown on the same figure are two "mean" ASME curves with T_0 determined by fitting the ASME equation to the K_a data for the 72W weld. The shift between the two curves is 84°C. It should be noted that the four duplex crack-arrest specimens were only irradiated to 1.56×10^{19} neutrons/cm² (> 1 MeV) compared to 1.88×10^{19} neutrons/cm² (> 1 MeV). No adjustment was made when they were considered as one set with the weld-embrittled specimens. Such adjustments may be made in the final report on the Sixth Series.

3.4 Preparations for Testing Irradiated Crack-Arrest Specimens Supplied by ENEA

A decision was made to build a larger crack-arrest fixture to accommodate the Italian specimens. The hot-cell fixture used in the hot cells with the Sixth Series (see above) is too small for the Italian specimens. The new fixture should also be capable of testing the other specimen sizes in use at ORNL. The design of the new fixture was based on the previous hot-cell fixture built and used at ORNL with the Sixth Series. Drawings were prepared and the manufacture of the components has begun. More details about this program are given below.

*All curve fitting methods rely on minimizing the residual between the fitted curve and the data in some manner. In this report, such curves are sometimes referred to as "mean" curves.

Figure 3.2 Crack-arrest toughness values for both unirradiated and irradiated 72W weld metal, and for weld-embrittled and duplex-type specimens.

The Italian Committee for Research and Development of Nuclear Energy and Alternative Energies (ENEA), the Italian NRC, started an extensive research program some time ago to characterize an ASTM A 508, class 3 forging produced in Italy. The research program encompassed both unirradiated and irradiated mechanical property data from the following types of specimens: tensile, Charpy impact (both standard and precracked), compact tensile, and crack-arrest. The testing and irradiation of these specimens was performed at several locations: ENEA CRE Casaccia Laboratories, Battelle Columbus Laboratories (BCL), and two laboratories of the French Commissariat à l'Energie Atomique. ENEA originally planned to test the irradiated crack-arrest specimens at BCL, but BCL has recently decommissioned their hot-cell facilities.

The NRC has agreed to test the irradiated crack-arrest specimens at ORNL because of the usefulness and applicability of these data to the safety assessment of U.S. RPVs. ENEA has nine irradiated crack-arrest specimens manufactured from the ASTM A 508, class 3 forging. The in-plane dimensions of three of the specimens are $25 \times 200 \times 200$ mm, and the remaining six are $13 \times 100 \times 100$ mm. The fluence of the nine specimens varied between approximately 2 to 3.2×10^{19} neutrons/cm² (> 1 MeV), and the irradiation temperature varied from 240 to 280°C.

The nine irradiated crack-arrest specimens belonging to the ENEA have been received from the Ford Nuclear Reactor in Michigan and are now stored at ORNL. The remote fixture used in the testing of the HSSI Sixth Irradiation Series is too small to be used with the three large specimens. There are other details about the ENEA specimens that have been discussed in the previous semiannual progress report. ORNL is in touch with BCL to keep them apprised of progress with the ENEA task.

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*Available in public technical libraries.

†Available for purchase from National Technical Information Service, Springfield, VA 22161.

4. Irradiation Effects on Cladding

F. M. Haggag

The objective of this series is to obtain toughness properties for two types of stainless steel cladding in the unirradiated and irradiated conditions. The properties obtained include tensile, CVN impact, and J-integral toughness. The goal is to evaluate the fracture resistance of irradiated weld metal cladding representative of that used in early pressurized-water reactors. The final report on the results of irradiation effects on three-wire cladding was published during the last reporting period.

During this reporting period, this task was inactive. However, a task to evaluate the effects of thermal aging on cladding is under way in the HSST Program, and the results are presented in HSST Program semiannual progress reports.

A limited number of previously precracked CVN impact specimens were included (December 1991) in the first large capsule of the HSSI Tenth Irradiation Series to obtain dynamic fracture toughness data on irradiated three-wire series-arc stainless steel cladding. These specimens are expected to be tested in 1994.

5. K_{Ic} and K_{Ia} Curve Shift and Annealing in LUS Welds

S. K. Iskander and R. K. Nanstad

The purpose of the Ninth Irradiation Series is to evaluate the correlation between fracture toughness and CVN impact energy during irradiation, annealing, and reirradiation (IAR). A preliminary review of the literature on the IAR response of RPV steels has indicated additional research needs. An important area identified is the rate of fracture toughness degradation due to neutron irradiation after annealing and, in particular, the relationship between fracture toughness and CVN impact energy.

In the next 2 years, IAR research in this task will utilize some of the substantial inventory of specimens irradiated in other HSSI Programs and stored at ORNL. The materials to be used have already been extensively characterized as part of the Second, Third, Fourth, and Fifth Irradiation Series. There are 120 so-called undersize* CVN specimens in this inventory, machined from HSSI weld 73W, which were irradiated in the spaces available in the Fifth Series capsules. A special container for annealing approximately 38 specimens at a time was designed and fabricated. The container will be placed in a controlled-atmosphere furnace that became available due to the cancellation of another program at ORNL. The temperature variation in the container was surveyed and found to be within $\pm 5^\circ\text{C}$ of the set point.

Annealing and testing of some of the undersize HSSI 73W weld CVN specimens is the first phase of this task. Approximately 90 CVN specimens from HSSI Welds 72W and 73W have been machined for inclusion in the Midland Capsule 10.5. These specimens will be annealed and, together with annealed undersized specimens, reirradiated to determine the rate of reembrittlement. This reembrittlement rate is one of the important parameters that determines the length of time a RPV can be operated safely after annealing.

A compilation of an IAR data base has been initiated. It has been used to evaluate published data on the IAR response of pressure-vessel materials irradiated at 260°C (500°F). Only seven data sets were found, but none on the reirradiation response. A similar search was performed by NRC, and their data were incorporated into the data base. The data base will continue to be updated. The data base contains 62 "parent" points, and approximately 360 data points related to IAR. At the request of NRC, the data base was transmitted to Modeling and Computer Services (E. Eason). Among the questions arising during the preparation of the data base was that the fluence reported was sometimes based on the equivalent fission spectrum and not the calculated fission spectrum. Moreover, data on the irradiation time, which are needed in many of the annealing response models, were often not available.

*The dimension normal to the notch is approximately 9.5 mm instead of the 10 mm of the standard full-size specimen.

6. Irradiation Effects in a Commercial LUS Weld

6.1 Chemical Composition and RT_{NDT} Determinations

(R. K. Nanstad, D. E. McCabe, R. L. Swain, and M. K. Miller)

The primary objective of the Tenth Irradiation Series is to investigate the postirradiation fracture toughness of the SAW from the Midland Unit 1 reactor vessel. The reactor is a PWR owned by Consumers Power Company and was canceled prior to startup. The weld from that vessel is of considerable interest because it carries the Babcock and Wilcox Co. (B&W) designation WF-70, an SAW fabricated with a specific heat of weld wire (heat 72105) and specific lot of flux (lot 8669). Welds with the WF-70 designation are contained in the beltline region of at least six operating nuclear reactor vessels and are the limiting materials (regarding irradiation effects) in at least two of those vessels.¹ The WF-70 weld was fabricated using copper-coated wire and Linde 80 flux and is known to be an LUS, high-copper weld. Major objectives of the unirradiated material characterization include determinations of copper variation, RT_{NDT}, and CVN upper-shelf energy throughout the welds. The results of those investigations have been completed and reported.² The following have been taken from that report:

Discussion

The B&W "Record of Filler Wire Qualification Test" shows results of CVN testing at 10°F (-12°C) as 35, 39, and 44 ft-lb (47, 53, and 60 J). Those results are somewhat above most of the curves reported here. The qualification record shows the qualification weld was postweld heat treated at 1100 to 1150°F (593 to 621°C) for 48 h. BAW-2070 reports that the Midland beltline weld received 22 h and 36 min at 1125 ± 25°F (607 ± 14°C), while the nozzle course weld received 25 h and 31 min at the same temperature. The slight differences between the weld qualification results would likely be mitigated if the Midland weld received a total of 48-h heat treatment. The shorter postweld heat treatment (PWHT) given the vessel welds would likely result in somewhat higher strength and higher transition temperature than that reported in the qualification tests and that is born out by the nozzle course weld, which has a higher yield strength than reported in the weld qualification, but is similar to that reported by Lowe³ for one WF-70 weld. It is not clear, however, why the beltline weld, which received a PWHT only 3 h less than that of the nozzle course weld, would have a yield strength significantly less than that of the nozzle course weld. This observation will be verified with tensile tests of the beltline weld.

The issue of RT_{NDT} determination is important for steels like the WF-70 weld because the RT_{NDT} is defined by the CVN toughness rather than the drop-weight NDT. The ASME Code is not perfectly clear as to the procedure to be used for such a determination. It requires the construction of a full Charpy curve from the minimum data points of all the C_V tests conducted. Strictly speaking, because Para. NB-2321 in the code defines a C_V test as three tested specimens, the above paragraph would require one to use the minimum value from each of those groups of three specimens. However, since it only requires C_V tests at NDT + 60°F or higher, one could not really develop a full C_V impact curve. On the other hand, the code allows determination of the RT_{NDT} when a C_V test has not been conducted at NDT + 60°F. It also is not clear how one should develop a curve from the minimum data points. It could be interpreted to be a best fit or an absolute lower bound. For consistency of RT_{NDT} determination, a clearer procedure needs to be developed and incorporated into relevant regulatory documents. Even from the analyses of the individual data sets, it is clear that substantial differences in RT_{NDT} can result depending on individual interpretation of the intended procedure. The data from the 7/8t (through thickness) location in beltline weld section 1-11 provide one example of this; Figure 6.1 shows the curve fits to all the data, to the minimum data as discussed previously, and a lower boundary curve. The lower boundary curve is about midway between the lower 68 and 95% confidence bounds. If the code paragraph is interpreted to mean the construction of a curve which encompasses all the data, the RT_{LB} for the data in Figure 6.1 would be about 29°C (84°F). That compares to an RT_{NDT} value of about 18°C (65°F) using the curve fit to the minimum data. The result depends, of course, on the actual construction of the lower boundary curve (e.g., smooth curve or point to point) as well, in that there is no guidance regarding its shape and, therefore, the 50-ft-lb (68-J) temperature could vary widely. For the individual beltline data sets, the RT_M ranged from 23°C (42°F) lower to 7°C (13°F) higher than the corresponding values of RT_{NDT}. For the individual nozzle course data sets, which generally had even fewer test results in each data set, there were no differences. A comparison of RT_{NDT} and RT_{LB} values shows that RT_{LB} ranged from 21°C (38°F) higher to 8°C (14°F) lower than the corresponding values of RT_{NDT}. Thus, the method used for determination of such an index is also dependent on the number of test results in the data set.

Figure 6.1. Comparison of various curve fits to the Charpy impact data from the 7/8 thickness location of Midland beltline weld sect.1-11. Shown are the hyperbolic tangent curve fits to all the data and to the minimum data points, a curve constructed as a lower boundary to all the data, and the 68 and 95% confidence bounds on the predicted value.

As seen in this study, small data sets for the same weld can result in substantial differences in determination of a transition temperature. The incorporation of statistical methods such as confidence bounds to be applied to the data would require a more well-defined prescription for the selection of test temperatures and number of tests. On the other hand, one could prescribe a value of deviation, based on documented behavior of welds and of base metals which could be added to the mean value determined from a curve fit to the data. The prescription must be chosen, of course, based on the desired confidence level, such as 95%. There are other potential methods, but they are dependent on various judgments that can result in significantly different results.

10CFR50 (ref. 4) states that a value of 0°F (-18°C) must be used as the RT_{NDT} for a Linde 80 weld if no credible measured value is available. For the three WF-70 welds referred to by Lowe,³ the RT_{NDT} values are 14, 23, and 51°C (58, 74, and 123°F, respectively). In this study of the Midland Unit 1 WF-70 welds, the RT_{NDT} (determined from a hyperbolic tangent curve fit to the minimum data points) varied from -20 to 35°C (-6 to 95°F). In SECY-82-465 (ref. 5), a standard deviation of 14°F (9°C) is given for Linde 80 welds. For the 25 values of RT_{NDT} in this study, the mean value is about 4°C (39°F) with a standard deviation of 14°C (25°F). Similarly, the mean value of upper-shelf energy is 88 J (65 ft-lb) with a standard deviation of 7.3 J (5.4 ft-lb). Using a curve fit to the minimum data for the entire Midland weld data set, the RT_{NDT} would be 15°C (59°F). Using the absolute lower bound, the RT_{LB} would be 35°C (95°F), about midway in the range reported by Lowe.³ Using a mean curve fit to the entire data set, the RT_M would be -3°C (27°F) with lower 68 and 95% confidence values of 18°C (64°F) and 67°C (153°F), respectively. The entire data set is shown in Figure 6.2. Thus, the temperature interval between the mean and lower confidence bound associated with one standard deviation on energy is about 21°C (37°F). The RT_{NDT} value of 15°C (59°F) is similar to the 68% confidence bound on the mean curve, 18°C (64°F). Similarly, the overall RT_{LB} value of 35°C (95°F) is close to the 90% confidence bound on the mean curve, 42°C (108°F). Regarding the 41-J (30 ft-lb) and 68-J (50 ft-lb) temperatures of -8°C (17°F) and 31°C (87°F), respectively, for the mean curve to the total data set, they are less than all the values reported by Lowe.³ His reported values, however, are within the 95% confidence bounds for the Midland data reported here.

Figure 6.2. Charpy V-notch impact energy versus test temperature for all tests of beltline and nozzle course welds of the Midland reactor vessel. The solid curve represents the mean hyperbolic tangent curve fit, while the dashed curves show a comparison of 95% confidence bounds on the mean and on the predicted value.

The fact that the RT_{NDT} values vary from -20 to 37°C (-3 to 99°F) warrants further consideration relative to both the unirradiated and irradiated test programs. The testing of multiple specimens to allow for statistical analyses is an integral part of the current planned program as is the testing and irradiation of fairly large specimens. The location in the weld from which specimens are machined will, of course, influence the unirradiated properties; however, because there was not a consistent trend of RT , 41-J temperature, or upper-shelf energy with depth in the welds (see Figure 6.3), one reference RT_{NDT} and 41-J temperature for the beltline weld and one of each for the nozzle weld will be used. It is very interesting that, despite the large scatter in these welds, the 95% confidence interval is similar to the relatively homogenous welds 72W and 73W (ref. 6) that were fabricated with copper contained in the weld wire and not as a coating as was used for the Midland welds.

The wide variation in copper content, however, will further complicate the analysis of radiation effects. This is particularly true regarding the differences between the beltline and nozzle course welds. Those differences will require the irradiation program to consider the welds as two different materials. Additionally, if 4T compact specimens are used in the program, the mechanical property and copper variations present in the weld will exist in individual specimens. The fact that this weld represents a real reactor vessel was the primary motivation for the study, yet the apparent variation in properties and copper content will only exacerbate the large variations already observed in relatively homogeneous materials. This does not necessarily render the study less useful, but it does require a different approach to the conduct of the program.

Figure 6.3. Plots of through-thickness Charpy impact results for the Midland beltline weld sections: (a) 41-J temperature, (b) upper-shelf energy, and (c) RT_{NDT}.

Observations and Conclusions

The major observations and conclusions from this study are as follows:

1. Drop-weight NDT temperatures from various through-thickness positions in the beltline and nozzle course welds varied from -40 to -60°C (-49 to -67°F).
2. The RT_{NDT} values (determined from a hyperbolic tangent fit to the minimum data in each case) in all 25 individual cases are controlled by the CVN impact energy and vary from -20 to 37°C (-4 to 99°F), with a mean value of about 4°C (39°F) and a standard deviation of 14°C (25°F). From a mean curve fit to the entire data set, the RT_M is -3°C (27°F) with a temperature interval between the mean and lower confidence bound associated with one standard deviation on energy of about 21°C (37°F). For the same 25 data sets, the RT_{LB} values (determined from the absolute lower bound in each case) vary from -21 to 35°C (-6 to 95°F) with a mean value of about 9°C (47°F) and a standard deviation of 16°C (29°F).
3. Analysis of the combined data from both beltline and nozzle course welds revealed a mean 41-J temperature of -8°C (17°F). Statistical analyses of the combined data (279 tests) show a 95% confidence temperature interval from -36 to 17°C (-32 to 62°F) at the 41-J energy level; that 53°C (94°F) interval width is similar to those observed for welds 72W and 73W in the HSSI Fifth Irradiation Series.
4. Analysis of the combined data set for the beltline and nozzle course welds (279 Charpy impact tests) gives a RT_M [50 ft-lb (68-J)] temperature from the mean curve fit less 60°F (33°C)] result of -3°C (27°F) with lower 68 and 95% confidence bounds of 18 and 67°C (64 and 153°F), respectively.
5. The ASME Code procedure for determination of RT_{NDT} is unclear, and different interpretations can result in substantially different results, depending on the nature of the data.
6. Upper-shelf energies for 25 data sets vary from 77 to 108 J (57 to 80 ft-lb) with a mean value of 88 J (65 ft-lb) and a standard deviation of 7.3 J (5.4 ft-lb). Analysis of the combined data from both beltline and nozzle course welds revealed a mean upper-shelf energy of 89 J (65 ft-lb) with 95% confidence bounds from 70 to 107 J (52 to 70 ft-lb).
7. Bulk copper contents range from 0.21 to 0.34% in the beltline weld (average is 0.26%) and from 0.37 to 0.46 wt % in the nozzle course weld (average is 0.40%); the overall average is 0.29% with a standard deviation of $\pm 0.07\%$.
8. Atom-probe field-ion microscopy (APFIM) revealed no evidence of detectable (darkly imaging) copper precipitates or clusters but did indicate substantial depletion of copper in the matrix over the bulk chemical analysis. Thus, copper precipitation occurred on a very fine scale as a result of the PWHT, but copper clustering did not occur.

6.2 Fracture Toughness of Unirradiated Material and Irradiation Program(D. E. McCabe, R. K. Nanstad, and R. L. Swain)

The activities of this reporting period involved finalization of the test matrix, machining and precracking of specimens, capsule building, and test equipment preparation. All compact, CVN, and tensile specimens have been machined and the precracking work has been completed. Special clip gages have been made for measuring load-line displacement from the top surfaces of specimens. Such gages were made for 1/2TC(T), 1TC(T), and 2TC(T) specimens.

There are six irradiation capsules, four of which are referred to as scoping capsules. Scoping capsules are dose- level studies, covering two materials (nozzle and beltline) and low-to-high irradiation levels of 5×10^{18} and 5×10^{19} neutrons/cm². Each capsule will have 14 CVN, 8 tensile, and 3 1/2TC(T) specimens. The status on scoping capsules is that we are currently evaluating a bid from Materials Engineering Associates to build and irradiate the two low-level irradiation capsules. This work should be completed this year. For the high-irradiation levels, the plan is to develop a small ORNL-developed capsule for this experiment and for use in future similar projects.

The principal irradiation objective is 1×10^{19} neutrons/cm², to be established in two large capsules (10.05 and 10.06). Capsule 10.05 is a design for 1TC(T) specimens. It contains the specimens shown in Table 6.1. The zone of best temperature control is dedicated to Midland WF-70 weld specimens. However, a few 1/2TC(T) specimens, coded 72W and 73W, are from the Fifth Irradiation Series. No 1/2TC(T) specimens had been included in that experiment, and this opportunity was taken to obtain additional data from surveillance capsule size specimens of that material. Table 6.2 lists the specimens that were positioned above and below the central zone of optimum temperature control. Temperatures can be expected to vary as much as 10°C away from aim control temperature in these regions. These specimens were added principally for postirradiation annealing experiments. There are twelve 1TC(T) Midland beltline specimens in this group. Dosimetry work was completed in April 1992, and capsule 10.05 is currently being exposed in the Ford Nuclear Reactor facility at the University of Michigan. Capsule 10.06 will be built this summer.

Currently, there are about 90 compact specimens and 24 tensile specimens that are to be tested for unirradiated material properties. This testing is currently under way.

Table 6.1 Capsule 10.05, specimens at preferred site

Material	Specimen type	Number of specimens	Levels of test temperature
MW beltline	1TC (T)	25	5
MW beltline	1/2TC(T)	24	3
72W	1/2TC(T)	8	1
73W	1/2TC(T)	8	1
MW beltline	Charpy V-notch	26	Transition curve
MW nozzle	Charpy V-notch	26	Transition curve
MW beltline	Precracked		
	Charpy V-notch	10	Transition curve
MW nozzle	Precracked		
	Charpy V-notch	10	Transition curve

Table 6.2. Capsule 10.05, specimens away from center

Material	Specimen type	Number of specimens
MW beltline	1TC(T)	12
MW beltline	Charpy V-notch	75
MW nozzle	Charpy V-notch	30
72W (Fifth Series)	Charpy V-notch	30
73W (Fifth Series)	Charpy V-notch	58
MW repair weld	Charpy V-notch	12
Clad metal	Charpy V-notch	18
Clad metal	Precracked Charpy V-notch	6
HSST Plate 02	Charpy V-notch	45
High Flux Isotope Reactor	Charpy V-notch	12
A 508	Charpy V-notch	12

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7. Microstructural Analysis of Radiation Effects

R. E. Stoller, K. Farrell, S. T. Mahmood,
M. K. Miller, and L. K. Mansur

7.1 APFIM Analysis of Midland Welds

The beltline weld of the Midland Unit 1 RPV was examined by APFIM. The APFIM analyses were conducted to characterize the nature of the precipitates, or clusters, especially those containing copper, in the microstructure and to compare the matrix copper content with the nominal bulk content. Similar APFIM analyses were conducted on the nozzle course weld and on both welds in the post-irradiation condition. High-quality field-ion micrographs of the weld were obtained. No evidence of darkly imaging copper precipitates or clusters were observed in any of the weld samples. Some ultrafine, brightly imaging refractory carbides was observed as shown in Figure 7.1. These carbides were observed in all five samples. The carbides were roughly spherical and approximately 1 nm in diameter. Similar ultrafine carbides have been observed in A302, grade B steel¹ and A533, grade B, class 1 steel.²

The average composition of each of the five beltline weld samples was determined in the ORNL energy-compensated atom probe, and the results are summarized in Table 7.1. The matrix copper content ranges from 0.07 to 0.13 wt %. The results indicate that the matrix copper content was substantially lower than the bulk value of 0.21 to 0.32 wt % obtained by chemical analysis. This behavior is consistent with other observations² and is due to copper precipitation at grain boundaries and other microstructural features during the PWHT.

The atom-probe composition data were subjected to statistical analysis to determine whether the matrix copper was in a random solid solution or if clustering had occurred. The data were analyzed to determine the significance of the mean copper atom separation and in terms of the Johnson and Klotz order parameter. In both cases, no deviation from a random solid solution was observed. Thus, although the matrix copper content was depleted due to the formation of fine copper precipitates, the remaining matrix copper had not clustered.

7.2 Modeling and Analysis

The embrittlement of pressure-vessel steels under irradiation is believed to be due to at least two types of defects. One is small, copper-rich precipitates, and the other is some sort of point defect cluster. This latter defect has been termed the matrix or radiation-induced defect and could consist of either interstitial or vacancy-type clusters. The modeling work initiated under this task is focused on examining the formation and evolution of these clusters and determining the degree to which these clusters contribute to radiation hardening. Irradiation temperature and displacement rate were identified as the two key variables to be explored. A detailed description of the model and the implications of the initial results obtained have been prepared.³

The major results of ref. 3 can be summarized as follows:

1. It appears that it is not appropriate to invoke the commonly used assumption of steady state point defect concentrations when modeling low-temperature embrittlement or when analyzing the results of low-temperature experiments. Using reasonable values for the necessary microstructural parameters, the time required for the point defects and the cluster populations to reach steady state was shown to be very long at low temperatures, e.g., it is on the order of 30 years at 60°C. The effect of displacement rate was observed to be different during the point defect transient and steady state regimes.
2. Although there is some uncertainty in determining the strength of point defect clusters as barriers to dislocation motion, results obtained using standard models from the literature indicate that both cluster types could induce similar levels of hardening. Using only the simplest barrier models, the calculated hardening was observed to be comparable to that observed experimentally. Since this work did not include any hardening contribution from radiation-induced precipitates, it appears that the cluster

Figure 7.1. Typical field-ion micrograph of Midland beltline weld section 1-13, region 4. An example of one of the ultrafine, brightly imaging refractory carbides is indicated by the arrow.

Table 7.1. Atom-probe analysis of the matrix composition for Midland reactor pressure vessel weld section 1-13

Weld region	Composition (wt %)									
	Cu	Ni	Mn	Mo	Cr	Co	Si	P	C	Fe
1	0.10	0.52	1.43	0.24	0.26	0.01	0.59	0.006	0.011	bal
2D	0.13	0.62	1.38	0.40	0.08	0.02	0.80	0.033	0.004	bal
2E	0.13	0.68	1.37	0.26	0.07	0.01	0.63	0.017	0.002	bal
3	0.07	0.48	1.21	0.62	0.17	0.04	0.81	0.028	0.035	bal
4	0.13	0.61	1.27	0.21	0.05	0.02	0.65	0.002	0.001	bal

contribution obtained from the model is probably somewhat too large. However, it seems reasonable to conclude that these clusters play an important role in hardening and that they may be responsible for greater hardening than the copper-rich precipitates for some conditions of dose, displacement rate, and temperature. An understanding of their behavior under thermal annealing may be particularly important for interpreting post-irradiation annealing studies and for predicting embrittlement under subsequent re-irradiation.

While many of the trends discussed in ref. 3 can be understood in terms of the various reactions between the point defect and the cluster populations, some of the details are not yet fully understood. Additional calculations have defined several areas that require further development to permit more confident comparisons between the model's predictions and experimental data. First, the range of uncertainty between plausible barrier models must be reduced. The ratio of the strengthening, calculated using a "strong" and a "weak" barrier model is shown in Figure 7.2 (refs. 3,4) as a function of the point defect cluster radius for two cluster densities. The interstitial clusters are assumed to be dislocation loops, and the vacancy clusters are assumed to be microvoids. Clearly, the uncertainty implied by Figure 7.2 is unacceptably large, particularly for the microvoids. An experiment is being investigated which may permit the direct measurement of the hardening increment due to small cavities. The experiment would involve the use of existing materials that have been implanted with helium and thermally aged to produce a range of bubble microstructures.

In addition to defining the hardening increment for any one type of defect (microvoids, loops, and precipitates), the method used to sum the contributions from the various components is also under investigation. Some guidance is available from calculations and proposed superposition laws in the literature (e.g., ref. 4), but further simple experiments are also being evaluated to provide a direct measurement for two-component microstructures.

One significant conclusion of this work is that embrittlement due to point defect clusters is not likely to increase at lower displacement rates compared to high displacement rates. As shown in Figure 7.3, the fraction of the point defects that survive matrix recombination does increase as the displacement rate is reduced. This is due to the competition between matrix recombination and sinks, such as dislocations. The matrix recombination rate is proportional to the product of the interstitial and vacancy concentrations while the rate of point defect absorption at sinks is proportional to either the interstitial or vacancy concentration alone. Thus, when the point defect concentration increases at higher displacement rates, the matrix recombination fraction increases approximately quadratically (the product of two increasing terms) while the sink absorption fraction increases only linearly.

Since the point defects that escape matrix recombination are those that can contribute to radiation-induced microstructural and property changes, this dependence of matrix recombination on displacement rate could give rise to a similar dependence for embrittlement. However, Figure 7.4 shows that the dependence of point-defect-cluster-induced strengthening on the displacement rate is opposite that of the matrix recombination fraction shown in Figure 7.3. At 60°C, the predicted strengthening at 0.1 (displacements per atom (dpa) exhibits little dependence on displacement rate. Most of the strengthening is due to clusters that are formed directly in the cascades. For a given dose, the same number of cascades will have been formed for any displacement rate. There is little diffusion of point defects at this temperature, so most of these clusters survive. The slight decrease in strengthening seen at lower displacement rates is due to some thermal annealing of the clusters over long times. The interstitial clusters show more annealing due to the lower interstitial migration energy. At 285°C, the annealing effect is much stronger. Interstitial clusters contribute little strengthening, and annealing of the vacancy clusters gives rise to reduced strengthening at lower displacement rates. Since the formation and evolution of the point defect clusters is not primarily controlled by point defect diffusion, the fact that a greater fraction of the point defects survive at lower displacement rates is of little importance to embrittlement by these clusters.

Some ongoing evaluation and development is indicated to verify the use of the current clustering model. The theory that has been used here was initially developed to investigate phenomena that occur at higher temperatures. The inability to use the steady state point defect concentrations under certain conditions was discussed above. This represents one example of the differences between modeling high- and low-temperature radiation effects, but there are others that require further examination. For example, the point defect and cluster densities that are predicted below 100°C (ref. 3) are high enough to potentially trigger a spontaneous, athermal recombination mechanism which would limit their further increase. Calculations have been reported which indicate that the recombination volume increases at lower temperatures and that the

Figure 7.2. Ratio of hardening values obtained with "strong" and "weak" barrier models for interstitial loops and voids.³

Figure 7.3. Influence of displacement rate on the fraction of interstitials that escape matrix recombination at 60 and 285°C.

recombination volume increases as the vacancy concentration increases. The implications of these calculations are being investigated to see if increased recombination would provide an upper limit to the point defect concentrations.

A new collaboration was initiated between ORNL staff and researchers at the Harwell Laboratory and the University of Liverpool in the United Kingdom. The purpose of this work is to investigate high-energy cascades in iron using the MOLDY molecular dynamics simulation (MDS) code^{5,6} and a newly developed interatomic potential for iron. While MDS studies have provided significant insight into primary defect formation and survival by permitting the behavior of displacement cascades to be studied in detail, most such studies have been conducted using interatomic potentials that are appropriate for face-centered cubic (fcc) copper.⁵⁻⁷ This work is intended to determine the degree to which the conclusions drawn from the fcc simulations can be applied to body-centered cubic (bcc) materials. Calculations of the first 10-keV cascade in iron have begun. Since the calculations require a great deal of computer time, current plans initially call for two cascades at this high-energy level. Other, lower energy cascades will also be simulated. Subsequent analysis and the availability of additional computer time will determine whether additional 10-keV cascades are done. The results of these MDS studies will provide additional guidance in the selection of damage parameters for the embrittlement model that is under development.

Figure 7.4. Displacement rate dependence of calculated strengthening due to interstitial and vacancy clusters at 0.1 dpa for irradiation temperatures of 60 and 285°C.

7.3 Low-Temperature Embrittlement

Several experimental studies of radiation-induced embrittlement in ferritic steels are supported in part under this task. The experiments are also supported by the Division of Materials Sciences at the U.S. Department of Energy (DOE). Part of the experimental work is being carried out in collaboration with researchers at the University of California at Santa Barbara (UCSB). The program at UCSB is funded by the Office of Energy Research in the U.S. DOE. These experiments are intended to examine the effects of neutron energy spectrum, neutron flux (atomic displacement rate), and alloy composition on low-temperature ($\sim 60^\circ\text{C}$) embrittlement. The experiments are being conducted in the High Flux Isotope Reactor (HFIR) at Oak Ridge, Tennessee, the High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory (BNL), and the Ford Nuclear Reactor at the University of Michigan.

Several engineering alloys are being irradiated in these experiments and include the HSST Plate-02 of A533B steel, an A36 steel of the type that is typically used in reactor support structures, an A588B steel obtained from the United Kingdom's Nuclear Installation Inspectorate, and the A212 and A350 archive materials from the HFIR. The material matrix includes a set of five simple model alloys (Fe, Fe-0.01 Cu, Fe-0.3 Cu, Fe-0.7 Ni, and Fe-0.3 Cu-) to investigate the influence of copper and nickel at low temperatures. More extensive compositional variation is provided by 18 simple model alloys and 32 model engineering alloys prepared by UCSB. Specimens include minitensiles of the ORNL SS3 type, coupons for microhardness and small-angle neutron scattering, and specimens for field-ion and transmission electron microscopy.

The neutron fluxes in the HFIR and HFBR are very high, 5×10^{18} and 5×10^{17} n/m²/s ($E > 1$ MeV), respectively, and the neutron exposures required to obtain measurable embrittlement are quite short (a few hours to a few days). The fluxes obtained in the Ford Nuclear Reactor irradiations are much lower, 2×10^{16} , 2×10^{15} , and 2×10^{14} n/m²/s. The use of these three reactors will provide a larger range of neutron fluxes than have previously been used in any single investigation of flux effects. Since the HFBR is a heavy-water-moderated reactor, the ratio of the thermal flux to the fast flux is very high, ~ 200 . In order to examine the influence of thermal neutrons,^{8,9} one set of the HFBR-irradiated specimens will be encapsulated in cadmium to eliminate thermal neutrons.

Some of the high flux irradiations in the HFIR hydraulic tube have been completed. As shown in Figure 7.5(a), for fluences less than about 5×10^{22} n/m², there is little systematic influence of copper and nickel on the yield strength data that have been obtained on the model alloys. At higher fluences, more strengthening is observed at higher copper and nickel concentrations. This result is similar to the behavior observed at higher temperatures^{10,11} and indicates that radiation-enhanced diffusion may lead to solute clustering even at 60°C . Little difference has been observed between the various engineering alloys in the HFIR; see Figure 7.5(b). The fluence dependent data shown in Figure 7.6 reflect the just completed dosimetry in the HFIR hydraulic tube.

Final preparations for the irradiation experiment at the University of Michigan Ford Nuclear Reactor have been completed. This experiment involves a collaboration between researchers at UCSB and ORNL. The irradiation conditions are summarized in Table 7.2. During this reporting period, dosimetry packets were prepared which contained wires of iron, nickel, titanium, copper, niobium, and an aluminum-1% cobalt alloy. The capsules also contained neptunium-237 and depleted uranium fission monitors. The wires and fission monitors were placed in a gadolinium vial to prevent thermal neutrons from influencing the results. An additional aluminum-1% cobalt wire was included in the irradiation capsules outside of the gadolinium to provide an estimate of the thermal flux. The irradiation capsules have been welded shut and will be shipped to Michigan early in October 1992 for insertion into the reactor.

Additional specimens were fabricated from several of the alloys used in the Michigan irradiations. These will be used in companion irradiations in the HFIR and the HFBR at the BNL. The purpose of these additional irradiations is to provide a comparison of neutron flux and spectral effects on a single set of materials. The fast neutron fluxes obtained in these three reactors will range from 2×10^{14} to 5×10^{18} n/cm²/s ($E > 0.1$ MeV). The use of highly thermalized locations in the HFBR will provide thermal-to-fast neutron ratios of greater than 200 at a fast flux of 4×10^{15} n/cm²/s. Companion irradiations will be conducted with thermal-to-fast ratios ranging from less than 1 to about 3. As mentioned above, several of the HFIR irradiations and dosimetry runs have been completed, and the data are undergoing further analysis. The HFBR irradiations should begin early in fiscal year 1993.

Figure 7.5. Increase in yield strength in (a) several model alloys and (b) engineering alloys following irradiation in the HFIR hydraulic tube. Fluences are only approximate at this time.

Figure 7.6. Yield strength as a function of fast fluence in several model and engineering alloys following irradiation in the HFIR hydraulic tube at $\sim 55^\circ\text{C}$.

Table 7.2. Irradiation matrix for joint UCSB/ORNL experiment in the University of Michigan Ford Nuclear Reactor^a

		Fast fluence (n/cm ²)			
		2 × 10 ¹⁷	5 × 10 ¹⁷	3 × 10 ¹⁸	2 × 10 ¹⁹
Capsule location and nominal fast flux (n/cm ² /s)	60D, 2 × 10 ¹⁰	-F1- 2778 h 115 d-18 h 161 d	-F4- 6944 h 289 d-8 h 406 d	n/a ^b	n/a
	60B, 2 × 10 ¹¹	-F-2 ^c 277.8 h 11 d-14 h 16 d	-F5- ^c 694.4 28 d-22 h 40 d	-F7- 4167 173 d-15 h 243 d	n/a
	60A, 2 × 10 ¹²	-F3- 27.78 h 1.15 d 1 d	-F6- 69.44 h 2 d-21 h 3 d	-F8- 416.7 17 d-9 h 24 d	-F9- 2778 h 115 d-18 h 161 d

^aThe upper line lists the required effective full-power hours (EFPH), the second line the effective full-power days (EFPD), and the bottom line lists the approximate number of calendar days based on a duty cycle of 10 days on and 4 days off.

^bA dummy capsule from ORNL, designated F0, will be run in the same sample holder as F1 and F4. The capsule will have an envelope of aluminum and stainless steel specimens in the external recess. The target fluence for this capsule is 5 × 10¹⁶ n/cm². This requires 694.4 EFPH or 28.9 EFPD, about 40.6 calendar days.

^cOn the initial loading, substitute capsules from ORNL will be used in place of F2 and F5. Irradiation Schedule - UCSB/ORNL Irradiation Experiment, University of Michigan, Ford Nuclear Reactor.

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8. In-Service Aged Material Evaluations

F. M. Haggag, R. K. Nanstad, and P. Arakawa

The overall objective of this task is to assess the service-induced degradation of fracture resistance through examination of components exposed during in-nuclear-plant operation. The initial focus of this task is to augment the existing hot-cell testing capability available to the HSSI Program with remote machining capabilities for the fabrication of specimens from samples of activated steel obtained from service-exposed components. During this reporting period, information and brochures were collected on electrodischarge machines and computer numerically controlled (CNC) machining centers suitable for hot-cell operations. A decision was made to pursue the acquisition of a CNC machining center. Three geometries of a 0.5TC(T), CVN, and flat tensile specimens were sent to the manufacturers of CNC machines for a bid to machine one each from carbon steel material to be provided by ORNL as the acceptance testing for the purchase of such a machine. The formal bid also stated that two ORNL engineers would witness the machining of the specimens and that specified tolerances should be achieved. Furthermore, the procedures and programming for machining these specimens should be provided with the delivery of the CNC machining by the successful bidder. Size requirements of the CNC machine were also specified in the purchase specification because of limited space available in the hot cells at ORNL.

9. Correlation Monitor Materials

W. R. Corwin

This task has been established with the explicit purpose of ensuring the continued availability of the pedigreed and extremely well-characterized material now required for inclusion in all additional and future surveillance capsules in commercial LWRs. Having recognized that the only remaining materials qualified for use as a correlation monitor in reactor surveillance capsules are the pieces remaining from the early HSST plates 01, 02, and 03, this task will provide for cataloging, archiving, and distributing the material on behalf of the NRC. The initial activities to be performed in this task will be to identify existing material and records in preparation of establishing a storage, monitoring, and disbursement facility. During this reporting period, a detailed inventory of all remaining correlation monitor material was completed.

As part of the detailed inventory of existing correlation monitor material, the outdoor storage yard for the materials stockpiled by the HSST Program was systematically examined. The positions of all pieces of steel were cataloged along with the identifying markings on each piece. Where the markings were still definitive, they were renewed. Where the markings had either weathered to the point of ambiguity or were not evident, as in the case of some material returned from early subcontract examinations, block sizing and other identifying marks were used to identify the material by cross-reference with existing records. Once the material remaining from HSST plates 01, 02, and 03, which all qualify as correlation monitor material, was definitively and completely inventoried, plans were initiated for it to be moved to a special HSSI archival storage facility. When the facility is completed, it will provide indoor storage for all properly pedigreed HSST plate 01, 02, and 03 material which represents the remainder of virtually all of the world's originally qualified correlation monitor material. The completed catalog of the portion of the steel inventory other than the correlation monitor material was provided to the HSST program staff for their use in subsequent material monitoring activities.

A piece of the correlation monitor material from plate HSST 02 was shipped to Professor A. Kumar at the University of Missouri, with NRC concurrence, for use in his NRC-sponsored research on subsized CVN specimens. The large body of irradiation effects data for the correlation monitor material will provide excellent benchmarks for evaluating irradiation embrittlement as measured with the subsized specimens.

10. Special Technical Assistance

This task has been established to explicitly emphasize and provide performance and financial monitoring of various analytical and experimental investigations conducted to support the NRC in resolving short-term, high-priority regulatory and research issues. The current activities being performed as part of this task include: providing expert guidance to the NRC staff regarding irradiation effects issues surrounding the embrittlement of the Yankee Reactor pressure vessel and providing dosimetry measurements for various surveillance specimen locations within the vessel of the HFIR at ORNL.

10.1 Yankee Vessel Integrity Assessments – R. K. Nanstad

The objective of this project was to provide expertise to the NRC staff regarding irradiation effects issues for the Yankee Reactor vessel and to perform analyses of the Yankee Atomic Electric company (YAEC) test reactor irradiation program with regard to determining the amount of embrittlement to the vessel. As part of those analyses, various consultations were held with the NRC staff on a number of different technical issues. Because the Yankee Reactor was shut down before completion of the YAEC test reactor program, a final evaluation of their test results could not be performed. As a result of requests for other pertinent analyses during the aforementioned consultation meetings, however, a number of letter reports were submitted. Two of those reports were co-authored by R. K. Nanstad and G. Robert Odette (UCSB) and are dated November 18, 1991, and January 20, 1992. In the letter report of November 18, 1991, the Yankee Reactor vessel embrittlement situation was evaluated, and various uncertainties were identified relative to that evaluation. Further evaluations regarding those uncertainties were performed and are discussed in the second letter report. That report discusses analyses of the power reactor embrittlement data base for margin analysis, copper/nickel comparisons, and comparison of transition temperature shifts as a function of specimen orientation. It also includes brief discussions of the surveillance data for the Yankee Reactor vessel upper plate and thermal annealing at 650°F. The third report, regarding statistical considerations in sampling of the Yankee Reactor vessel materials for analyses of chemical composition, was authored by R. K. Nanstad, K. O. Bowman, and D. J. Downing and was submitted in draft form to the NRC technical monitor for inclusion in a letter report to NRC.

10.2 Neutron Dosimetry Data for the HFIR Pressure Vessel – K. Farrell, F. B. Kam, C. A. Baldwin, W. R. Corwin, F. W. Stallman, L. Robinson, F. F. Dyer, and F. M. Haggag, and B. M. Oliver*

In February, 1992, a comprehensive neutron dosimetry package was irradiated in surveillance site Key 7, position 5, at the inside face of the pressure vessel of the HFIR at Oak Ridge, Tennessee. The primary goal was to measure the flux of thermal neutrons. Prior data on neutron fluxes and spectra at the surveillance sites had been obtained from calculations backed by measurements of stainless steel monitors in the surveillance packages, which yielded fast fluxes from activation of Ni and Fe in the alloy. Thermal fluxes were derived solely from neutron transport calculations that originally indicated a strongly thermalized spectrum with thermal (<0.4 eV)-to-fast(>1 MeV) flux ratios in the range 30 to 70 for positions in the Key 7 site. Newer calculations using improved codes and updated neutronics data cast doubt on the earlier estimates of thermal flux and showed that the spectrum at some locations on the vessel was not so thermalized; the new thermal-to-fast flux ratio at the Key 7 site was about 3. This first dosimetry experiment was intended to settle this discrepancy and to provide measured data as benchmarks for future calculations.

The Key 7 site was chosen for the experiment because it is the source of critical, low-temperature embrittlement data, and the surveillance package in position 5 could be temporarily removed without compromising the surveillance program. The monitors consisted of activation wires of Au, Ag, and Co for thermal neutrons and Ni and NpO₂ for fast neutrons, together with helium accumulation fluence monitors (HAFMs) of Al-Li and Al-B for

*Rockwell International.

thermal neutrons and Be for fast neutrons. Activities were counted at ORNL, and neutron reaction rates and fluxes were determined by ASTM standard procedures. The helium content of the HAFMs was measured at Rockwell International Corporation.

The results for thermal neutron flux show excellent agreement among the monitors. The values for the activation monitors range from 2.3 to $2.7 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. The values for the HAFMs are $2.26 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ for the Al-Li and $2.3 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ for the Al-B. The overall average value of $2.4 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ is about two-thirds of the recent calculated thermal flux of $3.7 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ for Key 7, position 5. Thus, these measured thermal fluxes not only establish the first reliable data for the HFIR vessel, but they also confirm the latest computed values. These conclusions are pertinent to the Key 7 site only; recent computations for some other surveillance locations on the HFIR vessel still indicate high thermal-to-fast flux ratios.¹

In this experiment, less attention was paid to fast flux because there has never been good reason to question the fast fluxes that are routinely measured from the stainless steel monitors in the surveillance capsules. They have always been of the expected order, and they agree reasonably well with neutron transport computations. The first comprehensive dosimetry experiment was seen as a chance to validate the earlier data with pure Ni monitors and as an opportunity to broaden the scope of the previous fast dosimetry by including for the first time the Np and Be monitors. Seizing that opportunity has proved to be a mixed blessing. Whereas the fast fluxes from the Ni monitors do, indeed, verify the previous measurements from stainless steel monitors, the data from the Np and the Be monitors are contentious.

Ni monitors were placed in two locations in the dosimetry capsule, one in a gadolinium vial with the Np monitor and a group of thermal neutron activation wires, and the other in an aluminum tube with thermal neutron wires at maximum distance from the gadolinium vial. The gadolinium-shielded Ni wire recorded a fast flux of $1.5 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. Previous measurements from Ni activation in stainless steel monitors had given a flux of $1.8 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$, and the recent calculations gave $1.2 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. In stark contrast to all of these values, the Np monitor yielded a fast flux of $2.6 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$, and the Be HAFM, which was located in a separate aluminum vial between the two Ni wires, indicated a flux of $5.8 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. A discrepancy of this magnitude between the Np and Ni monitors is very surprising, to say the least, and might be dismissed as human error if it were an isolated occurrence. But the second discrepancy (the Be HAFM), of the same order in the same direction but measured by different people with a different technique, is perhaps more than coincidental. We have searched for mistakes in our procedures, without avail. Another dosimetry experiment made in the HFIR core concurrently with the Key 7 dosimetry, using Ni, Np, and Be monitors from the same source materials, shows good agreement of the monitors, testifying to their veracity. We conclude that either there is a hidden fault in the Key 7 experiment or there is something exceptional about the moderated fast neutron spectrum at the Key 7 site. A new dosimetry experiment presently being assembled to monitor more of the surveillance sites on the HFIR vessel may help explain this mystery.

In short, the data from this first dosimetry experiment at the HFIR vessel Key 7 surveillance site gave a thermal neutron flux of $2.5 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$, in good correspondence with newer neutron transport calculations. Measurements of fast flux at $\sim 1.6 \times 10^{12} \text{ n}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ from Ni monitors agree with those from earlier stainless steel monitors and are close to the calculated values; measurements from Np and Be monitors give inexplicably higher fast fluxes.

Reference

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CONVERSION FACTORS^a

SI Unit	English unit	Factor
mm	in.	0.0393701
cm	in.	0.393701
m	ft	3.28084
m/s	ft/s	3.28084
kN	lb _f	224.809
kPa	psi	0.145038
MPa	ksi	0.145038
MPa•√m	ksi•√in.	0.910048
J	ft•lb	0.737562
K	°F or °R	1.8
kJ/m ²	in.-lb/in. ²	5.71015
W•m ⁻³ •K ⁻¹	Btu/h•ft ² •°F	1.176110
kg	lb	2.20462
kg/m ³	lb/in. ³	3.61273×10 ⁻⁵
mm/N	in./lb _f	0.175127
T(°F)=1.8(°C)+32		

^aMultiply SI quantity by given factor to obtain English quantity.

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11. ABSTRACT (200 words or less)

The primary goal of the Heavy-Section Steel Irradiation Program is to provide a thorough, quantitative assessment of the effects of neutron irradiation on the material behavior, and in particular the fracture toughness properties, of typical pressure vessel steels as they relate to light-water reactor pressure-vessel integrity. Effects of specimen size, material chemistry, product form and microstructure, irradiation fluence, flux, temperature and spectrum, and post-irradiation annealing are being examined on a wide range of fracture properties. The HSSI Program is arranged into 10 tasks: (1) program management, (2) K_{Ic} curve shift in high-copper welds, (3) K_{Ia} curve shift in high-copper welds, (4) irradiation effects on cladding, (5) K_{Ic} and K_{Ia} curve shifts in low upper-shelf welds, (6) irradiation effects in a commercial low upper-shelf weld, (7) microstructural analysis of irradiation effects, (8) in-service aged material evaluations, (9) correlation monitor materials, and (10) special technical assistance. This report provides an overview of the activities within each of these tasks from October 1991 to September 1992.

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