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J. A. Wang¹**DEVELOPMENT OF EMBRITTLEMENT PREDICTION MODELS FOR U.S. POWER REACTORS AND THE IMPACT OF THE HEAT-AFFECTED ZONE TO THERMAL ANNEALING**

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ABSTRACT: The NRC Regulatory Guide 1.99 (R.G. 1.99) Revision 2 was based on 177 surveillance data points and the EPRI data base, where 76% of 177 data points and 60% of EPRI data base were from Westinghouse's data. Therefore, other vendors' radiation environment may not be properly characterized by R.G. 1.99's prediction. To minimize scatter from the influences of the irradiation temperature, neutron energy spectrum, displacement rate, and plant operation procedures on embrittlement models, improved embrittlement models based on group data that have similar radiation environments and reactor design and operation criteria are examined. A total of 653 shift data points from the current PR-EDB, including 397 Westinghouse data, 93 B&W data, 57 CE data, and 106 GE data, are used. A nonlinear-least-squares fitting FORTRAN program, incorporating a Monte Carlo procedure with 35% and 10% uncertainty assigned to the fluence and shift data, respectively, was written for this study. In order to have the same adjusted fluence value for the weld and plate material in the same capsule, the Monte Carlo least-squares fitting procedure has the ability to adjust the fluence values while running the weld and plate formula simultaneously. Six chemical components, namely, copper, nickel, phosphorous, sulfur, manganese, and molybdenum, were considered in the development of the new embrittlement models. The overall percentage of reduction of the 2-sigma margins per ΔRT_{NDT} predicted by the new embrittlement models, compared to that of R.G. 1.99, for weld and base materials are 42% and 36%, respectively. Currently, the need for thermal annealing is seriously being considered for several A302B-type RPVs. From the macroscopic view point, even if base and weld materials were verified from mechanical tests to be fully recovered, the linking heat-affected zone (HAZ) material has not been properly characterized. Thus the final overall recovery will still be unknown. The great data scatter of the HAZ metals may be the result of the metallurgical heterogeneity that exists in the HAZ. The proposed data fitting procedure for the HAZ material is presented in the paper.

KEYWORDS: radiation embrittlement, power reactor, data base, LWR, reactor pressure vessel, embrittlement model, data fitting procedure, flux effect, heat-affected zone

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INTRODUCTION

Background

Aging and degradation of light-water-reactor pressure vessels is of particular concern because of their relevance to plant safety and the magnitude of the expected irradiation embrittlement. Irradiation embrittlement of RPV materials depends on many different factors such as neutron flux, fluence, neutron energy spectrum, irradiation temperature, and pre-irradiation material history and its chemical compositions [1-4]. These factors must be characterized in order to reliably predict the pressure vessel embrittlement and to ensure the safe operation of the reactor. Based on embrittlement predictions, decisions must be made concerning operating parameters, low-leakage-fuel management, possible life extension, and the need for annealing of the pressure vessel [5]. Large amounts of data obtained from surveillance capsules and test reactor experiments are needed. The data should comprise many different materials and different irradiation conditions to develop generally applicable damage prediction models that can be used for industry standards and regulatory guides. The embrittlement data base (EDB) is such a comprehensive collection of data for ferritic alloys resulting from the merging of the power reactor data base, PR-EDB [6], and the test reactor data base, TR-EDB [7]. There still is the unresolved issue of applying the test reactor data to the commercial power reactors' environment. Therefore, the current study is focused only on the PR-EDB data.

Heat-Affected Zone Materials

In general, reactor vessel embrittlement surveillance capsules contain materials, representing three conditions of the RPV steels, namely, base metal, weld metal, and heat-affected zone (HAZ). These three conditions may not share equal weight in the evaluation of radiation damage to the RPV steels. It has become a widespread practice in welding high-strength steels to incorporate a maximum HAZ hardness restriction in the fabrication specification in the offshore oil industry [8]. Heat-affected zones exhibited high hardness in the low alloy steels, and as hardness is increased the toughness generally decreases. What is important from a structural viewpoint is not the hardness per se but the fracture toughness of the HAZ relative to that of the adjacent base material. From the microstructure, material fabrication procedures, and macroscopic material behavior standpoints, the HAZ is indeed a very important factor for the determination of integrity [9-12]. Over 200 Charpy transition-temperature-shift data for HAZ metals are contained in PR-EDB. However, there is inconsistency in the data, as shown in Figs. 1-4. From Fig. 1, the HAZ impact test data has a large scatter band with low upper shelf, and the transition temperature falls between those for the base and the weld data. In contrast, the corresponding tensile test results, Fig. 2, indicate that the HAZ metal has the least ductility compared to base and weld metals. In Fig. 3, the HAZ impact data also seems to show that the transition temperature lower than for the base and weld metals, but the tensile data shows that the HAZ has the least ductility.

The great data scatter of the HAZ metals may be the result of the metallurgical heterogeneity that exists in the HAZ. In general, the HAZ contains four different regions, namely, coarse grain, grain refined, intercritical and subcritical regions, where the coarse-grained region is considered the most detrimental to the HAZ toughness. The HAZ data scatter may also due to the result of the inadequacy of conventional test techniques to measure the mechanical properties of

Fig. 1. Nine Mile Point One surveillance Charpy impact test results.

Fig. 2. Nine Mile Point One surveillance tensile test results.

Fig. 3. Palisades surveillance Charpy impact test results.

Fig. 4. Palisades surveillance tensile test results.

inhomogeneous/narrow/irregular geometry complex HAZ structures. Furthermore, in the absence of microstructure of sampling information, HAZ toughness data may be misleading.

The crack plane of the HAZ specimen has a tendency to branch into weld or base metals during a Charpy V-notch impact test, and does not always remain in the HAZ region [13]. This may be the direct result of difficulty in locating the notch tip accurately in the grain coarsened region of the HAZ specimens. Or due to the discontinuity nature of local brittle zones in the coarse-grained HAZ of multi-pass welds. Obtaining definite Cv toughness data in the coarse-grained region of the HAZ is difficult. The placement of a 0.254 mm radius notch in the correct location is at best hit and miss. The notch-tip radius is approximately the same size as the area of interest. Further the fusion line is not straight, which also contributes to the difficulty of notch placement. Even if the crack plane remained in the HAZ, different test results will be anticipated from coarse-grained or grain-refined regions. Nevertheless, when properly confining the crack plane of the Charpy impact test, for example by applying side groove on Charpy V-notch specimens, the uncertainty and data scatter of HAZ metal may be reduced.

Charpy Data Fitting Procedures

The current practice of Charpy curve-fitting procedures used in PR-EDB is based on the hyperbolic tangent model [14], which has been proven to be successful for the determination of transition temperature for materials which have more or less homogeneous properties. The impact energy and testing temperature are used as the two primary input parameters for the determination of the fitting curve. The hyperbolic tangent model relates impact energy E to the test temperature T according to the formula

$$E = \frac{(LSE + USE)}{2} + \frac{(USE - LSE)}{2} \cdot \tanh(SLOPE(T - TM)) \quad (1)$$

with USE and LSE the upper- and lower-shelf energy, respectively, with TM as the midpoint of the transition temperature region, and SLOPE as the slope of the curve at TM. This model is purely phenomenological but well characterizes the general shape of a Charpy curve in terms of the four basic parameters, USE, LSE, TM, and SLOPE. The hyperbolic tangent function is the most widely used fitting procedure next to hand (eyeball) fitting.

For a particular raw Charpy data set provided with chemistry information for each individual specimen, if certain data exhibit a large degree of scatter about the best-fit line, detailed study from PR-EDB [6] shows that these data generally either have a much higher copper or lower copper content than the rest of the specimens. This may imply that when the greater inhomogeneity of specific test sets are known, further constraints need to be added into the Charpy curve-fitting model, such as copper and nickel contents, to allow for material inhomogeneity. As for HAZ, an additional constraint, the average grain size of the fracture surface of individual broken specimen, needs to be considered in the final model fitting. In order to be able to use and analyze the surveillance HAZ data with better confidence, the following new hyperbolic tangent model for HAZ is proposed and described as follows:

$$E = \frac{(LSE+USE)}{2} + \frac{(USE-LSE)}{2} \cdot \tanh(SLOPE(T - TM)) + f(Cu, Ni, grain\ size) \quad (2)$$

where, f is a selected function, which contains input parameters Cu, Ni, grain size, and the corresponding constant fitting parameters. It can be written as follows:

$$f = A \cdot Cu + B \cdot \sqrt{Cu \cdot Ni} + C \cdot Ni + D \cdot (grain\ size) \quad (3)$$

where, A, B, C, and D are the constant fitting parameters. The best-fit equation of impact energy E

can be solved by using a nonlinear regression method to determine the constant coefficients of the selected model equation. The final index transition temperature can be obtained by substituting the index impact energy, mean chemistry, and mean grain size of the test set into the best-fit equation of impact energy E. Further information on chemistry compositions and grain size are needed to verify and quantify this HAZ model.

Within the near future, annealing recovery of RPV materials may be the number one option for mitigating radiation-induced embrittlement, thereby extending the life (beyond the 40-year nominal design) of RPVs in many nuclear power plants. Currently, the need for thermal annealing is seriously being considered for several A302B-type RPVs. The essential part of thermal annealing will be to quantify the percentage annealing recovery of HAZ. In author's opinion, the micro and macro material behavior of HAZ is a missing link. From the macroscopic view point, even if base and weld materials were verified from mechanical tests to be fully recovered, the linking HAZ material has not been properly characterized. Thus the final overall recovery will still be unknown. Also, due to the potential for thermal embrittlement, the overall beneficial outcome of thermal annealing on the RPV steels might be questionable. Initially, new RPVs are subjected to a 1150°F post-weld heat treatment (PWHT) to minimize cold cracking and residual stresses within HAZ and weld metals. The test reactor data shows generally that thermal annealing at 850°F for 168 hours can result in substantial recovery of RPV steel materials. However, there are two issues arising from these data, namely, (1) recent annealing data from the reconstituted specimens of Palisades surveillance specimens shows that only 65% recovery of the upper shelf was achieved after annealing at 850°F/168h [15], in contrast to 120% recovery of upper shelf as shown from the majority of the test reactor data (but it is not clear whether this is the result of the rate effect, or the result of the welding procedure used in manufacturing reconstituted specimen), (2) recovery of HAZ at 850°F has not been quantified. Thus, new thinking, innovative test techniques, and analytical methods need to be developed to answer the above questions.

Unfortunately, Charpy data scatter and the lack of post-irradiation annealing data for reactor pressure vessel steel HAZ materials limits the ability to accurately predict the response of HAZ to irradiation and to an 850°F anneal. The following new assessment of embrittlement data for base and weld materials suggests a route that might be applicable to HAZ material when more HAZ data is available.

PARAMETER STUDIES OF RADIATION EMBRITTLEMENT - BASE AND WELD MATERIALS

Irradiation Environment Effect

The degree of embrittlement is conventionally correlated with fast neutron fluence or with dpa, which are proportional to the production rate of point defects. However, radiation effects are driven not by the total atoms displaced but by the small fraction of point defects that avoid annihilation by mutual recombination and are made available to form clusters or to be absorbed at sinks [16-18]. Therefore, radiation effects are determined by the survival rate of point defects, not by their production rate. The survival rate of point defects (or defect availability) depends on several factors: (1) the temperature of the material, (2) the rate at which the fluence is accumulated, and (3) the energy of the neutrons causing the displacements. Thus the development of radiation embrittlement under different irradiation environments will not necessarily scale with fast fluence or with dpa unless the survival rate of point defects for each different irradiation scales in a similar way with the total defect production rate. Thus, from the embrittlement modeling point, data with large discrepancies in irradiation environment compared to others, may need to be separated from the main stream data, or to introduce additional constraints into the governing equations to control these variables, in order to generate

meaningful prediction models.

Neutron Flux Effect

Neutron flux effect is a well-known phenomenon in the test reactor environment, [14, 19]. From PR-EDB, neutron flux effect in power reactor embrittlement, in some extent, seems to indicate that neutron flux may also affect the embrittlement rate in the power reactor environment (see Figs 5-8). Based on PR-EDB, the plots of 30 ft-lb transition temperature shift (DTT30) vs. fast fluence were generated for base and weld materials, respectively. For this study, the chemistry contents were limited to copper content greater than 0.1 wt %, and nickel content no less than 0.5 wt %. Three ranges of neutron fluxes, namely, low flux, flux less than 10^{10} n/cm² s, intermediate flux, flux greater than 10^{10} n/cm² s and less than 10^{11} n/cm² s, and high flux, flux greater than 10^{11} n/cm² s, were investigated and identified with different symbols in the plots. In order to minimize the effect of the chemistry distributions on the trend curve, the plots were developed with normalized DTT30 (see Fig. 5, Fig. 7), where base metals were normalized with R.G. 1.99's chemistry factor (CF) equal to 137°F, and weld metals were normalized with CF equal to 145°F. The phenomena of the saturation of radiation embrittlement cannot be observed in Fig. 5 and Fig. 7. In Fig. 5, three trend curves were fitted for three ranges of neutron flux, respectively. The low flux data are all from General Electric (GE) boiling water reactor (BWR) data. From Fig. 5, the low flux data with fluence greater than $2 \cdot 10^{17}$ n/cm² are all located above the fitted trend curves of the intermediate flux and high flux data, in some extent, the trend curve of low flux data seems to indicate the possibility of the accelerated embrittlement for these low flux data; this is based on correlation of transition temperature shift with fast fluence. Also from Fig. 5, the trend curves seem to indicate that the embrittlement rate of high flux is lower than that of intermediate flux. The fitted trend curves on Fig. 7 also show that the embrittlement rate of high flux is less than the intermediate flux for weld materials. Further investigation was done on the same data set, but excluding the B&W's data to minimize the impact of irradiation temperature on the data trend, see Fig. 6 and Fig. 8. Again, the mean trend curve seems to indicate that the flux effect may exist in power reactor environments. The above mentioned flux effect is embedded into the development of the new embrittlement models implicitly by separating vendor specific data. Also further subdivisions, based on power output, were also applied to Westinghouse data.

Irradiation Temperature Effect

Irradiation temperature undoubtedly exerts a strong influence on embrittlement by controlling the migration rate of vacancies, the bulk recombination rate, and the annealing rate of clusters. Therefore, without proper consideration of irradiation temperature effect, bias will result in the embrittlement model.

A residual study on transition temperature shift was carried out, based on Version 2 of PR-EDB. Where the residual is defined as measured shift minus R.G. 1.99, Rev. 2's prediction. The residual study clearly indicated a large bias appeared for Babcock & Wilcox (B&W) surveillance data. The residual is defined as the reported transition temperature shift value minus the predicted shift of R.G. 1.99, Rev. 2. The mean residual and its corresponding 2-sigma uncertainty, do not take into account any absolute residual value greater than 100°F (there are 11 such observations, which are from Yankee Rowe and Garigliano), and were -4.51°F and 58°F for the base materials, respectively; and -3.04°F and 68°F for the weld materials, respectively. Detailed residual studies were done for each individual vendor: Westinghouse, GE, B&W, Combustion Engineering (CE), and Allis-Chalmers. The mean residual and corresponding 2-sigma uncertainty of base and weld for each individual vendor are listed in Table 1.

Fig. 5. Normalized shift for base metals with Cu>0.1, Ni>=0.5 wt%.

Fig. 7. Normalized shift for weld metals with Cu>0.1, Ni>=0.5 wt%.

Fig. 6. Normalized shift (w/o B&W) for base with Cu>0.1, Ni >=0.5 wt%.

Fig. 8. Normalized shift (w/o B&W) for weld with Cu>0.1, Ni >=0.5 wt%.

TABLE 1--The mean residual and 2-sigma of plates and welds per vendor data.

Vendor	Base Residual °F			Weld Residual °F		
	mean	2-sigma	points	mean	2-sigma	points
Westinghouse	-2.43	51°F	253	-0.85	61°F	93
B&W	-23.40	59°F	55	-23.43	64°F	23
GE	-0.04	77°F	50	-2.10	76°F	42
CE	-2.33	34°F	30	4.57	84°F	14
Allis-Chalmers	33.00	56°F	6	11.75	20°F	4
Overall	-4.51	58°F	394	-3.04	68°F	176

A clear bias was shown in the B&W data, and it indicated that R.G. 1.99, Rev. 2, overestimated the shift. One explanation is that B&W's reactors are operated at a higher temperature (the nominal capsule temperature of B&W is about 608°F from melt wires) than reactors of other vendors.

The melt wires of a surveillance capsule indicate the maximum temperature of an irradiated capsule, but a transient effect may compromise the accuracy of this type of thermal monitor. Due to the uncertainty of the irradiation temperature from the surveillance reports, an index irradiation temperature was determined and defined as "If both maximum and minimum capsule temperature were given, which were based on melt wires, the average is taken as the index irradiation temperature. Otherwise, use whatever is available." Average irradiation temperature of each individual vendor is listed in Table 2.

TABLE 2--The mean irradiation temperature of PR-EDB per vendor.

Vendor	Mean irradiation temperature	2-sigma	Data point
Westinghouse	578°F	30°F	353
B&W	608°F	22°F	80
CE	554°F	10°F	45
GE	554°F	24°F	101

Table 2 indicates that surveillance specimens experienced different irradiation temperatures with different vendors, which implies that the temperature effect needs to be considered in embrittlement modeling.

The temperature experienced in a given irradiation capsule is determined by the local coolant temperature, the gamma heating level, and the thermal resistance between specimen and coolant. The capsule temperature (or the specimen's temperature) may deviate somewhat from that of the coolant inlet temperature, depending on the irradiation environment and capsule design, and this situation may be magnified for a capsule which has high thermal resistance (i.e., there is a gap between the specimens and the capsule).

In the B&W capsules, significant melting of the thermal monitors is reported consistently, and, while this may well be due to transients, as stated in the reports, significantly higher irradiation temperatures cannot be ruled out. The melt wires from 15 B&W capsules indicated that irradiation temperatures were greater than 610°F. The overall mean index irradiation temperature for B&W's capsules is 608°F from melt wires. While from B&W's mean coolant inlet temperature data, most of the coolant inlet temperatures are below 558°F, this makes up to a 50°F difference as compared to 610°F (index irradiation temperature from melt wires). The transient phenomena alone may not be sufficient to explain and quantify this difference, and further investigation on the quantification of the irradiation temperature of surveillance specimens may be needed.

Different reactor designs operate with different average capsule temperatures, based either on inlet water temperature or melt-wire temperatures, but the precise average temperature is uncertain, so it makes sense to separately consider the data from the different vendors.

Material Parameters

Because of the different production process, the base and weld metals physically present different metallurgical structures and their pre-existing defects. Thus, in the current study, the base and weld metals were modeled with different equations in the embrittlement modeling

Analysis of A302B and A533B reactor pressure vessel steels in the surveillance capsule of commercial power reactors [14], seems to indicate that, from the measured transition temperature shift point the embrittlement behavior of both the A302B and A533B-1 plate materials is very similar. Therefore, no further modeling parameters were made to distinguish A302B and A533B materials.

From past studies [20] on "sensitivity of specimen orientation to radiation embrittlement" from the statistical point, there is no difference in transition temperature shift of TL vs LT orientations. Thus, no distinction was made between TL and LT data in the embrittlement modeling. And on "the phosphorus effect," the results seem to indicate that the phosphorous effect appears with low-copper content materials [20].

Background Study for Reg. Guide 1.99, Rev. 2

The R. G. 1.99, Rev.2 is the starting point for the development of this new embrittlement models. Thus, this historical note is very important because it reveals where new models come from and why these new models are needed. Three major data bases and two embrittlement models were involved in the modeling of R.G. 1.99, Rev. 2 [21]. The three data bases are R.G.'s 177 data points, EPRI's data base [22], and French test reactor data [23,24]. The content of the data base and the impact of its related model to R.G. 1.99, Rev. 2, are stated in the following paragraphs.

George Guthrie's model, 177 data points: A total of 51 weld and 126 base metal data points were taken from surveillance reports by Randall and rechecked by Guthrie. These data were used in Guthrie's embrittlement model [25]. Of the 177 data points, 128 fluences can be identified as Simons' data, which are registered in NUREG/CR-3319. The chemistry data are consistent with that of PR-EDB. Twenty-five shifts can be identified as 50-ft-lb shifts, and 146 shifts can be identified as 30-ft-lb shifts. The fluence factor (FF) of R.G. 1.99, Rev. 2, is mainly determined by Guthrie's FF with slight modifications. For base metal, Guthrie's chemistry factor (CF) is used with two exceptions. First, when Guthrie's CF for base metal exceeded those for welds predicted by Odette, the latter were used. Second, at very low copper levels, the CF is assigned as 20°F. The 2-sigma margins of R.G. 1.99, Rev. 2, is based on the residual study of the 177 data points.

Odette's model, EPRI data base: The EPRI data base used in Odette's modeling [26], which contains 65 weld and 151 base metal data, was collected directly from surveillance reports, except for the shift value which is determined by EPRI's hyperbolic tangent fitting procedure. EPRI's data base overlapped 177 data points almost completely. The fluence values are the same for 97% of the surveillance's fluence data. The chemistry data are consistent with that of PR-EDB. For weld materials, Odette's CF is used as the main guideline for R.G. 1.99, Rev. 2, except with very low copper a minimum value of 20°F is used.

French test reactor data: Since the U.S. surveillance data base had very few data for low copper steel, French test reactor data was used to provide guidance. The French test reactor data contain 56 base and 21 weld data which are collected from refs. 23 and 24. For both base materials with copper less than 0.03% and weld with copper less than 0.01%, the CF was assigned as 20°F.

DEVELOPMENT OF NEW EMBRITTLEMENT PREDICTION MODELS

Regarding the microstructure defect evolution, all residual defects created through the radiation events will contribute to the final macroscopic material property changes. The residual defects in materials due to neutron induced-displacement damage are a function of neutron energy, neutron flux, and exposure temperature, as well as the material properties that determine how neutrons interact with atoms and how defects interact within the material. Thus, "temperature, neutron flux, neutron energy spectrum, and material composition and processing history" all contribute to the radiation embrittlement. Insufficient considerations of the above-mentioned conditions may result in misleading correlation and thus incorrect prediction of material state and material behavior, as well as incorrect end-of-life determinations.

In general, the major contributions to the margin of the embrittlement model come from the inherent scatter in mechanical test results, neutron fluence evaluation, irradiation temperature, chemical analysis, and inhomogeneous material properties of the sample specimens. Past studies [27] on "Westinghouse's improved fluence data and its impact on 2-sigma margins" show that insufficient consideration of all the above-mentioned error sources in the determination of the correlation procedure will not result in substantial improvement of the margin of the embrittlement model. The margins and model evolution of the current R.G. 1.99, Rev. 2, are based on Randall's 177 data points and EPRI's data base, where 76% of 177 data points and 60% of EPRI's data base were from Westinghouse; therefore, the other vendors' radiation environment may not be properly characterized in the Regulatory Guide's prediction model. The above statement can be verified from the mean residual and 2-sigma margins of each individual vendor listed in Table 1. In particular the mean residual for B&W is significantly lower than the other vendors. In Table 1 the overall 2-sigma margins of weld and base materials per PR-EDB, Ver. 2, are 68°F and 58°F, respectively. Both margins are worse than that of R.G. 1.99, Rev. 2, 56°F and 34°F for weld and base materials, respectively.

Some factors are not well characterized, either due to lack of information, or data with large uncertainty, or incompleteness of damage mechanism model, such as topics related to irradiation temperature, neutron flux, neutron energy spectrum, and the final residual defects. For example, as stated in "parameter study" section, a large uncertainty exists in the index irradiation temperature of melt wires, and the option of using the coolant inlet temperature as irradiation temperature for the surveillance specimens may need further justifications. Clearly, a complete and accurate description of microstructural development during irradiation has not been attempted and seems to be beyond the present state of art.

It is proposed here that one way to tackle the problem of dealing with insufficient information, and the lack of complete damage mechanism modeling on irradiation temperature, neutron flux, and neutron energy spectrum effects, is to classify the embrittlement data into groups that have similar radiation environments (which imply that the reactors have similar design and operating procedures). This way, more realistic embrittlement models with reduction of the margins should be generated.

From the point of view of reactor design, surveillance capsule designs, reactor operating procedures (including irradiation temperature, heat-up and cool-down procedures, and core fuel loading configuration), and the sufficiency of the available surveillance data for the regression analysis, it was decided to group the data per individual vendor (B&W, CE, GE, and Westinghouse). The Westinghouse surveillance data was further divided into three subgroups, based on the power reactor thermal output.

In the development of R.G. 1.99, Rev. 2 [25], Guthrie had shown that conventional (zero error

assumed in fluence values) least square fitting procedures underestimate the value of the fluence factor. And the Monte Carlo method was used in Guthrie's study by assuming a 35% standard deviation in fluence and a 15°F standard deviation in Charpy measurements. The standard deviation of fluence can be varied from 20% to 35% depending on the specific weight used in Guthrie's least square fitting routines. A 35% upper bound uncertainty is assigned to the fluence data in the new models may well be appropriate for the fluence listed in the power reactor surveillance reports. This 35% may be due to the errors derived from dosimetry measurements, transport calculation, cross section data, and inconsistency among the surveillance reports regarding to choosing of neutron dosimetry for the final fluence evaluation. For example, some reports may choose either iron monitors, copper, or nickel, or they may average all of the dosimetry data to determine the final fluence value.

A total of 653 shift data points from the current PR-EDB, limited to U.S. power reactor data, including 397 Westinghouse data, 93 B&W data, 57 CE data, and 106 GE data, were used in the current embrittlement model development. A FORTRAN program was written for this study, using the IMSL nonlinear-least-squares fitting subroutine ZXSSQ, and incorporated with a Monte Carlo procedure with 35% and 10% uncertainty (1 standard deviation) assigned to the fluence and shift data, respectively. The uncertainty assigned to the fluence and Charpy measurement is used to simulate and reflect the error sources from the fluence evaluation and mechanical test results. Since no proper estimate on uncertainty of chemical composition can be determined, thus no uncertainty was assigned to chemistry data in the current study. In order to have the same adjusted fluence value for the weld and plate material in the same capsule, this program has the capability of adjusting the fluence values while running the weld and plate formulas simultaneously.

The author used the average specimen chemistry obtained from the broken Charpy specimens as the primary input for the model fitting whenever the data is available; otherwise, manufacturers' chemistry is used in the model fitting. In such a way the particular set of Charpy impact tests can be closely related to its chemistry compositions

The surveillance report's shift is used as input for the embrittlement modeling. However, if there is a large discrepancy between the surveillance report's shift and EDB hyperbolic tangent shift, the surveillance report's data is used as input unless engineering judgment indicated that the EDB fit is more appropriate.

New embrittlement prediction equations:

The development of new prediction equations stem from a series of studies on radiation embrittlement models, such as Guthrie's model [25], Odette's model [26], Fisher's model [28], B&W Lowe's model [29], the French FIM model [30], etc., and several other parameter studies. Although the copper-precipitation model has been extremely successful in explaining many aspects of irradiation embrittlement, it is becoming increasingly evident that other elements also contribute to the embrittlement of the RPV steel, such as Ni, P, Mn, Mo, and S. Theoretically, all the impurities in the low alloy steel are candidates to be included in the modeling. For example, C, Si, etc., were investigated in the test run, but including or excluding these elements did not affect the overall outcome of the statistical parameters significantly; therefore, these parameters (or elements) were not incorporated into final governing equations. Also due to the lack of the completeness of the available chemistry data from PR-EDB, Cu, Ni, P, Mn, Mo, and S were tentatively selected as key chemistries and were incorporated into the formula of the new prediction equations. Furthermore, the reason for separating weld and base metals is because the weld presents a possible region of physical and metallurgical discontinuity. The welding process offers added chances for the introduction of defects

and undesirable components or stresses.

The development of the parameters for this new embrittlement models is based on statistical formulation chosen by computer iterations, in some extent, the physical mechanism is embedded in the equations, such as the formulation of the fluence factor. The new prediction equations for each individual vendor are listed below:

Westinghouse Model 1 (thermal power output < 2000MW) :

$$\begin{aligned} \text{Shift(Weld)} &= [C(1) \cdot Cu + C(2) \cdot \frac{\sqrt{Cu \cdot Ni}}{Mn} + C(3) \cdot \frac{P}{Cu}] [f^{C(4)} \cdot C(5) \cdot \ln f] \\ \text{Shift(Base)} &= [\frac{C(6)}{Mn} + C(7) \cdot Cu + C(8) \cdot \sqrt{Cu \cdot Ni} + C(9) \cdot P] [f^{C(10)} \cdot C(11) \cdot \ln f] \end{aligned} \quad (4)$$

Westinghouse Model 2 (2000 ≤ thermal power output ≤ 3025MW) :

$$\begin{aligned} \text{Shift(Weld)} &= [C(1) \cdot Cu + C(2) \cdot \sqrt{Cu \cdot Ni} + C(3) \cdot Ni] [f^{C(4)} \cdot C(5) \cdot \ln f] \\ \text{Shift(Base)} &= [\frac{C(6)}{Mn} + C(7) \cdot Cu + C(8) \cdot \sqrt{Cu \cdot Ni} + C(9) \cdot S] [f^{C(10)} \cdot C(11) \cdot \ln f] \end{aligned} \quad (5)$$

Westinghouse Model 3 (thermal power output > 3025MW) :

$$\begin{aligned} \text{Shift(Weld)} &= [C(1) \cdot Cu + C(2) \cdot \sqrt{Cu \cdot Ni} + C(3) \cdot Ni] [f^{C(4)} \cdot C(5) \cdot \ln f] \\ \text{Shift(Base)} &= [\frac{C(6)}{Mn} + C(7) \cdot Cu + C(8) \cdot \frac{\sqrt{Cu \cdot Ni}}{Mn} + C(9) \cdot P] [f^{C(10)} \cdot C(11) \cdot \ln f] \end{aligned} \quad (6)$$

Babcock & Wilcox:

$$\begin{aligned} \text{Shift(Weld)} &= [C(1) \cdot \frac{Cu}{Mn} + C(2) \cdot \frac{\sqrt{Cu \cdot Ni}}{Mn} + C(3) \cdot S] [f^{C(4)} \cdot C(5) \cdot \ln f] \\ \text{Shift(Base)} &= [\frac{C(6)}{Mn} + C(7) \cdot Cu + C(8) \cdot \sqrt{Cu \cdot Ni} \cdot Mo + C(9) \cdot Ni] [f^{C(10)} \cdot C(11) \cdot \ln f] \end{aligned} \quad (7)$$

Combustion Engineering:

$$\begin{aligned} \text{Shift(Weld)} &= [C(1) \cdot Cu + C(2) \cdot \sqrt{Cu \cdot Ni} + C(3) \cdot S] [f^{C(4)} \cdot C(5) \cdot \ln f] \\ \text{Shift(Base)} &= [C(6) + C(7) \cdot Cu + C(8) \cdot \sqrt{Cu \cdot Ni} + C(9) \cdot P] [f^{C(10)} \cdot C(11) \cdot \ln f] \end{aligned} \quad (8)$$

General Electric:

$$\begin{aligned} \text{Shift(Weld)} &= [C(1) \cdot Cu + C(2) \cdot \sqrt{Cu \cdot Ni} \cdot Mo + C(3) \cdot S] [f^{C(4)} \cdot C(5) \cdot \ln f] \\ \text{Shift(Base)} &= [C(6) + C(7) \cdot Cu + C(8) \cdot \sqrt{Cu \cdot Ni} \cdot Mo + C(9) \cdot \frac{P}{Cu}] [f^{C(10)} \cdot C(11) \cdot \ln f] \end{aligned} \quad (9)$$

where fluence, f , is in units of 10^{19} n/cm² ($E > 1$ MeV), and Cu, Ni, P, S, Mn, and Mo are in wt %. Constant coefficients $C(I)$, $I = 1$ to 11, are fitting parameters, and are listed in Table 3.

TABLE 3—Fitting parameters per new embrittlement equations

Vendor	C1	C2	C3	C4	C5	C6	C7	C8	C9	C10	C11
B&W	-150	527	4261	.239	-.0611	19.3	1560	-1446	93.9	.2041	-.0632
CE	197	410	-1763	.083	.0327	-55.9	166.	433	1532	.3556	-.0585
GE	164	819	-1052	.294	-.0141	-112	394	659	611	.2801	.00439
W_1	61.5	549	114	.217	.0796	-27.6	472	283	-1486	.1807	.02802
W_2	-324	887	-125	.202	-.0110	-28.0	763	144	-2176	.3238	.02921
W_3	193	513	-82	.114	-.1282	-57.5	636	122	2568	.1813	-.1093

The resulting 1-standard deviation of residuals for each individual vendor are compared with that of R.G. 1.99, Rev. 2 in Table 4. Absolute residual values greater than 100°F are not accounted for in these results. The 1-standard deviation of residuals calculated by the new embrittlement prediction models, were based on the unadjusted surveillance fluence, and unadjusted average specimen chemistry, and unadjusted measured shift (either from reported or hyperbolic tangent fit data) from PR-EDB. The adjusted values were only used during the models development. The final estimate of the two sigma bounds, was purely based on the reported unadjusted data. All the 2-sigma margins for the new equations have a significant improvement compared with the Regulatory Guide's margin. The overall 2-sigma margins for the weld and base materials of the new model are 35.26°F and 31.02°F, respectively. The 2-sigma margins for the weld and base materials of Reg. Guide 1.99, Rev. 2, are 60.60°F and 48.30°F, respectively. The overall percentage of improvement of 2-sigma margins per ΔRT_{NDT} for weld and base materials are 42% and 36%, respectively. The data trends for the Regulatory Guide model and new models are demonstrated in the residual plots; less data scatter was revealed in the plots of the new model than in the plots of the Regulatory Guide model (see Figs. 9-12).

TABLE 4—Comparison of 1 standard deviation of residual for plate and weld.¹

Vendor	Reg. Guide 1.99		New Model	
	Residual °F (point)		Residual °F (point)	
	weld	plate	weld	plate
B&W	39.23 (31)	29.86 (62)	17.07 (31)	13.83 (62)
CE	30.88 (18)	21.00 (36)	10.45 (19)	10.53 (38)
GE	35.60 (39)	37.33 (67)	20.30 (39)	17.72 (67)
Westinghouse	25.25 (107)	18.88 (290)	17.97 (107)	15.91 (290) ²
			W_1 15.24 (24)	16.24 (79)
			W_2 15.28 (31)	17.06 (88)
			W_3 20.72 (52)	14.95 (123)
Overall	30.30 (195)	24.15 (455)	17.63 (196)	15.51 (457)

¹Comparison was made based on PR-EDB data.

²The 1 standard deviation of Westinghouse's is based on residual from W_1, W_2, and W_3.

CONCLUSION

Improvement in the 2-sigma margins is achieved by taking into account properly grouped data and adequate chemistry compositions into the fitting formula. By properly grouping data with similar reactor design and operating procedures, former differences in radiation environments, that induce

Fig. 9. R.G. 1.99 residual for surveillance base materials.

Fig. 10. New residual for surveillance base materials.

Fig. 11. R.G. 1.99's residual for surveillance weld materials.

Fig. 12. New residual for surveillance weld materials.

effects of spectrum, flux, and temperature are minimized, resulting in substantial improvement in 2-sigma margins. the impact of these parameters on the embrittlement modeling.

Currently, dpa is used as a measure of the potential to create point defects. It is not necessarily equal to, or even proportional to, the actual number of residual point defects. However, a better correlation parameter for radiation embrittlement can be expected through the upgraded dpa that incorporated the concepts of residual defects. Clearly, a complete and accurate description of microstructural development during irradiation has not been attempted and seems to be beyond the present state of art. Thus, before the real damage mechanisms can be fully understood, and the real embrittlement models are fully developed, this new embrittlement models can service as an intermediate tool for the investigation of regulatory related issues, and as a calibration and benchmark means for the future development of the more sophisticate embrittlement prediction models.

Continued needs exist for expanded data bases on materials typical of current practice and from various sources of supply, and for continued development and standardization of test methods and techniques for both surveillance and post-irradiation testing procedures.

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