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A PLANT-SPECIFIC UPPER SHELF ENERGY DROP METHODOLOGY

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ABSTRACT: Appendix G to 10 CFR 50 requires that licensees evaluate all reactor beltline materials to determine whether the Upper Shelf Energy (USE) screening criterion will be exceeded prior to the end-of-license (EOL). Measurement of the unirradiated USE and estimation of the EOL irradiated USE level are key elements of the screening criterion evaluation. The current Regulatory Guide 1.99 (Revision 2) USE drop model does not provide adequate representation of the actual shelf drop behavior for some reactor materials. Therefore, research was conducted to identify the important factors which should be included in the development of a physically-based USE trend curve model and to test the validity of the material-specific modelling approach.

As a result of this investigation, it has been concluded that there are three radiation damage mechanisms which may have significant influence on the ductile fracture process: precipitation and particle coarsening due to radiation enhanced diffusivity; cascade induced matrix damage; and possibly surface active element transport to grain boundaries and particle interfaces. At present, it is believed that the largest micromechanical effect is due to the impact of precipitates and matrix damage on the hardening of the material bridges between voids. However, additional experimental data are needed to confirm the dominant mechanisms which contribute to the USE drop.

In the absence of an experimentally verified microstructural model, work has been performed to test the efficacy of the material-specific approach. The validity of the material-specific approach has been demonstrated by reducing the Light Water Reactor (LWR) database to produce a data set with comparable chemical composition, heat treatment, and neutron flux spectra. Based on the work reported herein, it has been observed that a drop in the unirradiated USE requires an incubation dose. The reduced data set correlates well with the square root of the dose, indicating the importance of the fine scale precipitates and the matrix damage component.

KEYWORDS: Upper Shelf Energy, trend curve, ductile fracture, neutron damage, hardening, embrittlement

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LOW UPPER SHELF ENERGY ISSUE

Appendix G to 10 CRF 50 requires a minimum Upper Shelf Energy (USE) of 68 J throughout the operating life of the plant. Operation below the USE screening criterion is allowed if it can by shown that the lower USE provides sufficient margins of safety against fracture equivalent to those margins specified in Appendix G of the ASME Code. An elastic-plastic fracture mechanics assessment can be performed to determine the lowest USE which can be licensed. Recent work has shown that USE levels as low as 31 J (for a Boiling Water Reactor (BWR)) can be justified on the basis of elastic-plastic fracture mechanics calculations [1, 2]. In addition to the screening criterion, Appendix G to the ASME Code requires all plates and forgings to be tested in the T-L orientation (many surveillance specimens are L-T orientation). NRC Branch Technical Position MTEB 5-2 states that the L-T values may be reduced to 65 % of their original value to yield T-L properties. Application of MTEB 5-2 to the lowest irradiated datum yields an USE near the screening criterion for many older plants.

MATERIAL-SPECIFIC METHODOLOGY

Nine Mile Point Unit 1 began commercial operation in 1969 and, as with most older plants, low and/or dropping USE values pose ongoing challenges for continued operation. The Regulatory Guide 1.99 (Revision 2)(RG 1.99(2)) model indicated that the beltline plate materials would fall below 68 J prior to EOL, but, data from the first two surveillance capsules indicated no shelf drop. The need to better understand the material-specific behavior of the reactor pressure vessel materials at Nine Mile Point Unit 1 became evident.

As stated in the ASME Code, Section XI, Article A-4400, it is intended that plant-specific fracture toughness data be determined directly in the surveillance program. In particular, the ASME code states, "Radiation induced changes in fracture toughness should be determined from surveillance specimens of the actual material and product form, irradiated according to the surveillance techniques of ASTM E 185, ..." Since most surveillance programs do not include fracture toughness specimens, the effects of neutron irradiation are considered for both K_{IA} and K_{IC} by shifting the reference nil ductility temperature (RT_{NDT}) as a function of irradiation using trend curves. In particular, the generic RG 1.99(2) models are used to determine the Charpy shift at the 41-Joule level (ΔT_{41}) and USE drop (ΔUSE) due to neutron damage. In cases where two or more credible surveillance data are available, the Regulatory Guide provides a procedure for combining the measured data with the generic model to predict the ΔT_{41} and ΔUSE fluence dependence. As stated in ASME Code Section III, Article A-4400, this approach is intended to be extremely conservative: "These curves (the shifted K_{IA} and K_{IC} curves) are intended to be very conservative since the recommended procedure is to determine the irradiation effects from surveillance specimens of the actual material and product form in question."

As a result of the conservatism inherent in generic approaches and the complexity of the phenomenology, it is highly desirable to develop material-specific and plant-specific models. This can be achieved by using miniature specimen technology to increase the amount of data obtainable from current surveillance programs [3,4]. Miniature specimen technology, together with surveillance capsule re-insertion technology [5], will eventually provide sufficient data for development of plant-specific trend curves. In the meantime, material-specific models can be developed which are based on surveillance data for a particular plant combined with data from other plants with vessels fabricated using similar materials.

EXISTING MODELS

The current RG 1.99(2) model, which is identical to the 1977 Revision 1 model, proposes the following functional form:

$$\Delta USE(\%) \propto C(Cu)(\text{fluence})^{0.14}$$

where, C(Cu) = function of Cu content.

Other researchers have extended hardening theories used to explain Charpy shift and yield strength increase to account for the observed USE drop (see Table 1). No one has addressed the problem of relating the neutron induced microstructural evolution to the ductile fracture process to produce a physically based ΔUSE trend curve. Along these lines, there are several important questions concerning RPV steels exposed to neutron irradiation which must be addressed before a physically-based model can be developed:

- Do 1-5 nm defect clusters/precipitates influence the ductile fracture process by hardening the ligaments (ductile bridges) between voids and possibly cause flow localization?
- What are the levels at which yield strength elevation and strain hardening cause significant changes in the elastic-plastic response of the ductile bridges?
- At what surface concentrations does phosphorous (and other surface active elements) segregation to grain boundaries and particle interfaces result in an intergranular or particle/matrix delamination fracture component on the upper shelf?

MICROSTRUCTURAL/COMPOSITIONAL CONSIDERATIONS

Steels for Reactor Pressure Vessel (RPV) plates, such as A302M and A533B, are 0.23C steels with about 1.35Mn, 0.5Ni and 0.5Mo (weight percents). In addition, their levels of impurities or tramp elements are generally 0.01-0.02P, 0.01-0.02S and 0.1-0.2Cu [6]. These impurities can have dramatic effects on their mechanical behavior depending in part on the processing of the steels. To produce the heavy plate needed for RPVs, the steels are generally cast into large ingots which, after cooling, are hot rolled or forged into thick plates which are then re-austenitized, quenched, and tempered. The plates are then welded into the vessel and the final structure is stress-relief annealed and furnace cooled.

As a result of this processing, the plate, as it is put into service, is neither chemically nor microstructurally homogeneous. During the slow cooling of the large ingot through the solidification range, macrosegregation of alloying elements and impurities occurs which produces chemical and microstructural banding and stringers of non-metallic inclusions (sulfides, oxides, oxy-sulfides) throughout the thickness of the plate after it has been forged or rolled. The re-austenitization at 840-900°C and quenching of the plates during heat treatment produces a primarily bainitic structure. The subsequent tempering at about 660°C has several effects: it anneals out some dislocations which causes softening; it causes precipitation of a variety of carbides (e.g., initially, coarse Fe_3C which causes softening, and later fine Mo_2C which has a strengthening effect); it coverts some retained austenite to ferrite plus carbide.

TABLE 1--Summary of ΔUSE trend curve correlations

Author	Reference	ΔUSE Equation (%)[1]
Kass, et al[2]	6	$\Delta USE = C_2 + C_1 \left[(\%Cu) \left(\frac{\phi t}{10^{19}}\right)^{\frac{1}{3}} \right]$ or $\Delta USE = C_2 + [C_1(\%Cu) + C_3(\%P)] \left(\frac{\phi t}{10^{19}}\right)^{\frac{1}{3}}$
Varsik, et al	7	$\Delta USE = [C_1 + C_2(\%Cu) + C_3(\%Si)] \left(\frac{\phi t}{10^{19}}\right)^{C_4}$
Odette	8	$\Delta USE = (C_1 \log[C_2(\%Cu, \%Ni)] - C_3) \left(\frac{mdpa}{30}\right)^{C_4}$
NRC	9	$\Delta USE = C_1(\%Cu) \left(\frac{\phi t}{10^{19}}\right)^{C_2}$
Gutherie	10	$USD = UUS^{C_1} \{C_2 + C_3 \ (EL)\} \ (\phi t)^{C_4 + C_5 \ln(\phi t)}$
Pachur	11, 12	$\Delta USE = \Delta\infty(x,\phi) \ f(\phi) \ f(T) \ [1 - e^{-C_1 \phi t}]$

[1] ϕ = fast flux n/cm²/sec
t = time
C_i = regression constants
$\Delta\infty$ = saturated relative USE drop
$f(\phi)$ = flux effect function
T = irradiation temperature
x = (%Cu) [%Ni + %Mn]
UUS = unirradiated USE
USD = upper shelf drop
EL = possible important elements

[2] Same equation used for 30 ft-lb transition temperature shift.

The final stress-relief anneal of the welded vessel at about 630°C, is in fact a further tempering, so that Mo_2C precipitation and coarsening continues, and there is a further reduction of retained austenite. The final furnace cooling of this heavy vessel is extremely slow (several days) which raises the possibility that the copper, which at levels of Cu<0.3% is in solution at the tempering/stress-relief temperatures, will precipitate. However, thermal aging studies of similar steels cited by [13] indicate that at levels of Cu<0.3% there is insufficient supersaturation at thermal aging temperatures of 450-550°C to precipitate the Cu. At the lower temperatures, where the supersaturation increases, the diffusivity of Cu in Fe is too low to support precipitation. However, during the slow furnace cooling through the range 575-375°C, there is the danger that the segregation of impurities such as P, As, Sb and Sn, if present in sufficient quantity, may result in temper embrittlement. Many RPV steels meet the compositional requirement for temper embrittlement, (%Mn+%Si)(%P+%Sn)≥ 0.02, [14], and Miller and Burke [15] have reported enrichment of P at grain boundaries and carbide/matrix interfaces in stress relieved A533B welds.

During service, as these plates are exposed to irradiation, there is an increase in σ_y, and an upward shift in T_{41}. These changes have been shown to correlate with Cu concentrations [16,17], and since there are numerous observations of Cu-rich defects in the irradiated material, the conclusion has been drawn that these defects are responsible for the changes in mechanical behavior [13,15]. There has also been observed a correlation between ΔUSE and $\Delta\sigma_y$ [18,19] which may or may not imply a relation between Cu-rich defects and USE drop. However, property changes (especially ΔUSE) caused by other metallurgical reactions (carbide formation, P segregation, and change in retained austenite) have received much less attention, and the extent to which they contribute in RPV steels is largely unknown. There is a long history of metallurgical studies of these phenomena by means of thermal aging (see for example any physical metallurgy text, e.g., Reed-Hill). These microstructural changes can be easily envisioned to occur since neutron-induced nucleation sites are created and the vacancy supersaturation accelerates diffusional transport.

Accordingly, a study of the effects of thermal aging on mechanical properties was begun using an RPV steel in which Cu precipitation would not play a significant role (that is a steel with Cu<0.3%), but in which the other significant metallurgical reactions may occur (that is, a steel in which there are sufficient Mn, Si, and P to induce temper embrittlement, sufficient Mo to produce Mo_2C, and significant amounts of retained austenite). The steel examined was a modified A302B, designated G-8-3, which had been quenched from 850°C, tempered for four hours at 660°C and air cooled, and finally stress relieved for 40 hours at 620°C and furnaced cooled. Its composition is as follows:

C	Mn	P	S	Si	Cu	Ni	Mo
.23	1.13	.023	.018	.16	.17	.59	.48

The aging response of the hardness of this steel was determined by isochronal anneals (one hour) in the temperature range 100 to 650°C. The results (Fig. 1) show an initial decrease in hardness (ΔHV=35) between room temperature and 400°C, which is indicative of the commonly observed first stages of tempering. In this range, softening results from precipitation and growth of carbides of iron which is a consequence of the high diffusivity of interstitial carbon. Between 400 and 600°C, where self and substitutional-solute diffusivities become significant, there is a significant hardness increase (ΔHV=23) which is normally attributed to secondary hardening caused by dissolution of some Fe_3C to form the more stable Mo_2C at temperatures where Mo becomes mobile. However, there may also be a contribution from the transformation of the retained austenite to one or more austenite decomposition products by means of Fe self diffusivity.

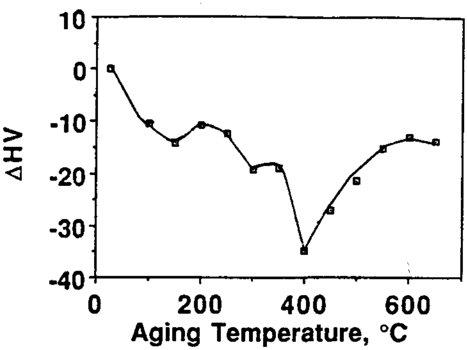

FIG. 1--Aging response of G-8-3 to isochronal (one hour) anneals

To examine this hardness peak in detail, the response of G-8-3 to isothermal anneals was examined in specimens which were first pre-aged at 300°C for eight hours to remove the initial low-temperature hardness drop. Aging curves at 500 and 550°C are shown in Fig. 2. At both temperatures, hardness increases from HV 187 to ~210 occur in periods of two to three hours. Other temperatures are being examined to determine the activation energy of the hardening process. These are significant hardness increases corresponding to an increase in tensile strength of this steel from 600 to 670 MPa. Since little Cu precipitation is expected, this hardening is believed to be caused by the transformation of retained austenite or formation of Mo_2C. Specimens in the underaged, peak-aged, and over-aged condition are being examined by TEM and XRD for changes in precipitate structure and retained austenite. Initial results of the TEM examinations indicate that there has been a marked reduction in retained austenite as a result of the aging treatment. Quantitative data on the reduction in retained austenite are forthcoming from XRD studies. Since there was very little change in the USE between the as-received and peak aged conditions, we have concluded the P segregation and modest hardening have little effect on the USE for this steel. Future neutron irradiation studies are planned to quantify the effects of Cu precipitation and other microstructural changes on the USE.

Candidate Mechanisms

From a phenonemological perspective, there is evidence that hardening mechanisms play a role in shelf drop. Reference [18] proposed a non-linear correlation between fractional decrease in USE (f) and increase in yield strength. Although the data set used in the mid-1980's suggested a possible nonlinear dependence of f with increase in $\Delta\sigma_y$, Figure 3 shows that a linear dependence is more appropriate. The proposed linear function is:

$$f = 2.18\times10^{-3}\ \Delta\sigma_y \tag{1}$$

Based on the data shown in Figure 3, it appears that Cu precipitation, carbide precipitation, and/or matrix damage play an important role in lowering the USE. Future work must focus on the reason for the apparent correlation between strength increases and shelf drop. It is not certain at present whether the ductility loss implied by yield strength elevation is the cause of ΔUSE or perhaps the major effect is the flow localization caused by the shearable fine scale defects. However, as shown in Figure 4, the USE in the irradiated and unirradiated condition correlate strongly with Reduction of Area (RA) as expected for the ductile fracture process. Since the change in RA (ΔRA) due to irradiation is typically ~10% (or less) of the unirradiated RA level, the ΔUSE and ΔRA do not correlate well.

We hypothesize, in concert with Broek [20], that the large particles (>1μm) (of relatively low number density) are stable under irradiation and fail early in the fracture process (ex. MnS). Since these particles fail at low strains, they cannot be essential to the irradiated material fracture process other than lowering the initial ductility of the material. Intermediate size particles (~500 - few thousand Å) are expected to initiate voids which grow by slip and thereby control the ductile fracture process. RPV steels contain about 3% by volume of carbide particles which range in size from 0.05 to 0.5 μm in diameter. The interfaces of these carbides with the matrix are the primary nucleation sites for the microvoids that ultimately grow and link to cause ductile failure. This nucleation may be unchanged by irradiation, however, the growth mechanism of the microvoids is critically dependent upon the stress state, the work hardening behavior of the material, and the distribution of carbide initiation sites. Examination of the Reference [16] data base suggests only modest changes in the work hardening exponent after irradiation. The reason for this is that in the irradiated steel, vacancy clusters, collapsed loops, and Cu clusters/precipitates are formed which act as shearable obstacles to dislocation motion. Rather than diffusing the strain by slip-plane hardening, as is the case with nondeformable particles which leave dislocation debris on the slip plane, shearable obstacles cause

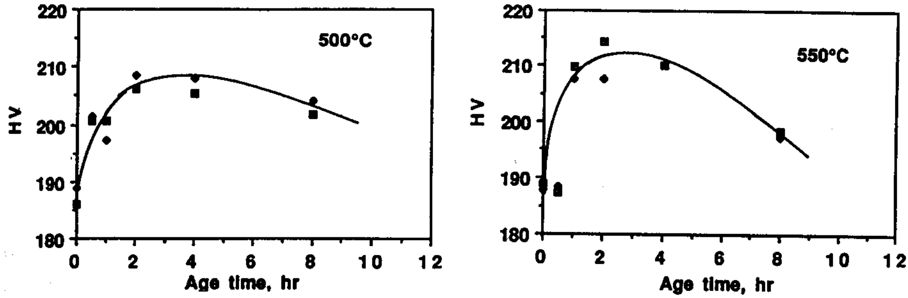

FIG. 2--Aging response of G-8-3 to isothermal (500 and 550°C) anneals.

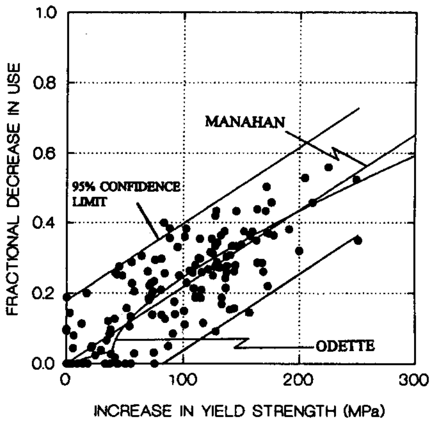

FIG. 3--Yield strength model based on a linear correlation between fractional decrease in USE vs. $\Delta\sigma_y$ (data from PR-EDB)

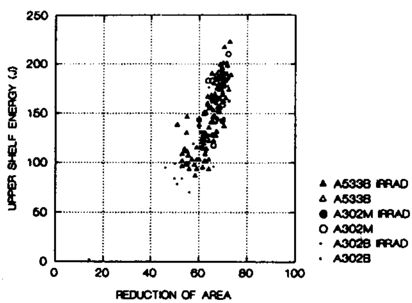

FIG. 4--Correlation between upper shelf energy and reduction of area (reference [16] data).

localization of strain in the unhardened slip plane [21] Such strain localization in the ligaments between microvoids would be expected to lead to ligament failure by the flow localization mechanism of void linkage [22] which in turn results in premature ductile failure.

A substantial amount of research will be required in the future to determine the extent of non-hardening embrittlement (ex., P transport to surfaces) in RPV steels. McElroy [23] has recently shown that P can be transported to grain boundary interfaces during final heat treatment and can have a significant effect on ductile fracture. McElroy [23] has also discussed grain boundary embrittlement in higher P Russian steels and MAGNOX welds. However, it is important to bear in mind that intergranular fracture has not been reported in the U.S. steels. This is most likely because the concentration of surface active elements at boundaries has not reached a critical level for fluences in the low 10^{19} n/cm^2 range.

MATERIAL-SPECIFIC MODEL

Baseline data on the effects of compositional variables on the USE of RPV steels have been analyzed by Manahan and Cuddy [24]. The steels had been quenched and tempered, and finally stress relieved at 625°C and furnace cooled. These data are of interest because this latter treatment could have been sufficient to allow some precipitation and/or segregation to have occurred prior to irradiation. Of the elements reported in the LWR database, only increases in Mn, S, P, and Cu increase the drop in USE (Figure 5). Presumably these correlations result from precipitation of Mo_2C and Cu, the segregation of P (and possibly Cu and S) to internal interfaces and grain boundaries, and the formation of S containing inclusions (e.g., MnS), all of which will promote loss of ductility. It also appears that the beneficial effects of Ni addition can be offset by high S content as shown in Figure 5.

In the absence of an experimentally verified, physically-based ΔUSE trend curve model, a material-specific methodology has been developed and applied to the Nine Mile Point Unit 1 (NMP-1) reactor. The procedure accounts for hardening and non-hardening mechanisms including:

1) Carbide and Cu rich precipitation and coarsening due to enhanced diffusivity of minor alloying and tramp elements

2) Cascade induced matrix damage influence on the ductile fracture process

3) P (and other surface active elements) transport to grain boundaries and to particle interfaces

In order to investigate the importance of the variables identified, and to test the validity of the materials-specific approach, the Power Reactor-Embrittlement Database (PR-EDB) was subdivided such that the final data set is representative of the Nine Mile Point Unit 1 (NMP-1) plates. A plot of ΔUSE vs. fluence for the entire database analyzed is given in Figure 6. Examination of the unirradiated USE and ΔUSE as a function of various chemical elements and fluence suggests that the A508 and A302B plates should be modelled as separate sub-populations. Therefore, the A302M and A533B data were treated as one data set and subdivided into a chemistry range which is representative of the NMP-1 beltline plates. The key characteristics of the NMP-1 material-specific model are as follows:

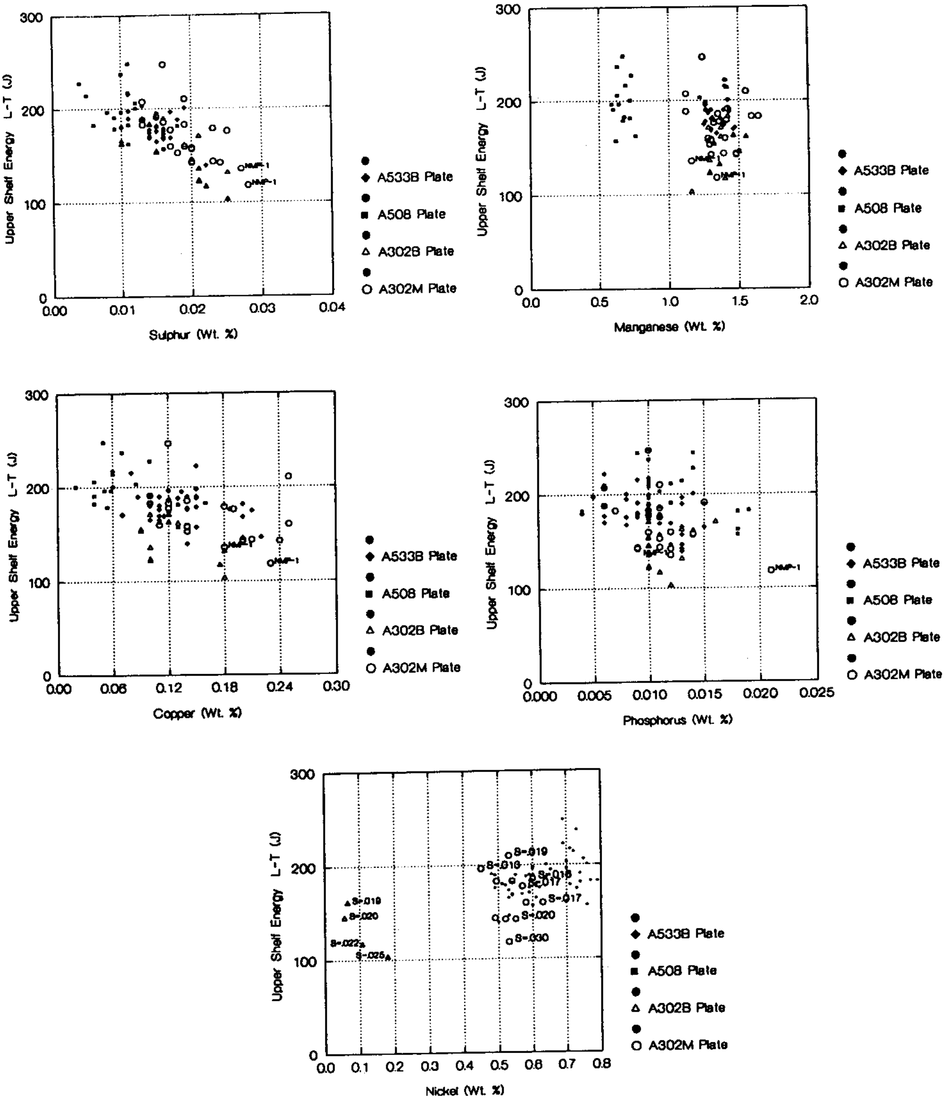

FIG. 5--The effects of various elements on the USE of RPV steels.

- Only power reactor data are included.
- Since the A302B ΔUSE sensitivity is much lower than that of the A302M and A533B materials, the A302B data were dropped. (As a result of high S content, the A302B materials have low unirradiated USE levels and relatively small ΔUSE with fluence.)
- As a result of low Mn and S levels, A508 data were dropped. The remaining plates have Mu in the range $1.1 \leq Mn \leq 1.65$ and sulfur in the range $.01 \leq S \leq .028$.
- Only plates of similar heat treatment were included (quenched and tempered)
- Only plates with C in the range $0.18 \leq C \leq 0.22$ were included
- Only plates with Cu in the range $0.18 \leq Cu < 0.28$ were included
- Only plates with Ni greater than 0.45 were Included
- Only plates with P in the range $0.010 \leq P \leq 0.021$ were included

For small and/or soft particles and depleted zones, dislocations tend to cut through without looping. A key parameter is the volume fraction (f) of the dispersed particles. References [25, 26] suggest that the change in yield strength should be proportional to the square root of volume fraction. Since volume fraction is proportional to dose, one would expect to see correlation when plotted versus square root of dose. Since intergranular fracture has not been reported for irradiated U.S. steels, it has been assumed that non-hardening embrittlement can be ignored. However, we recognize that radiation enhanced diffisivity is sufficient to transport surface active elements to grain boundaries and particle interfaces. While grain boundaries have not reached critical containment levels, it is recognized that additional work is needed to characterize effects on particle delamination with the matrix.

A regression analysis was performed and the results are shown in Figure 7. It is important to note that the 2σ confidence intervals for the model shown in Figure 7 are on the order of the experimental uncertainty for measuring the USE on the upper shelf. The resulting equation is listed below:

$$\Delta USE = -4.09 + 1.22\ (\text{fluence})^{1/2}/10^{8} \quad (J) \tag{2}$$

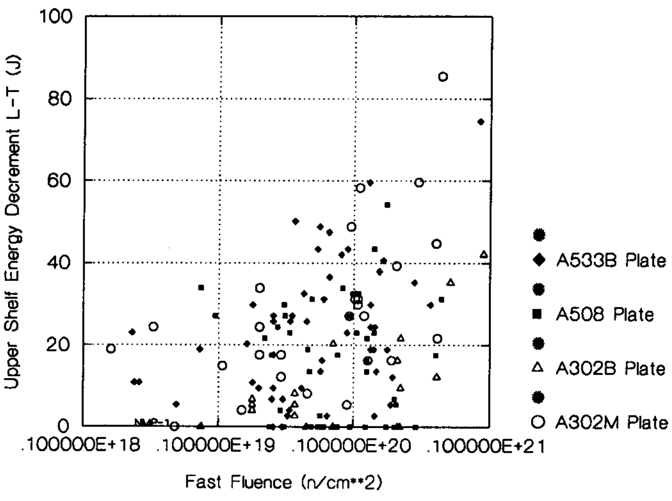

FIG. 6--ΔUSE vs. fluence for LWR vessel materials

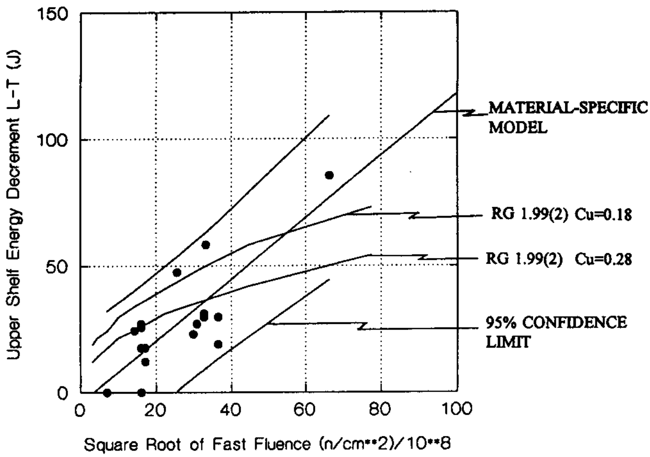

FIG. 7--Material-specific model for A302M plate

In practice, the model described above will be fit to actual plant data as more data become available. For NMP-1, the ΔUSE ~ 0 for a fast fluence of 4.78×10^{17} n/cm^2. Therefore, the incubation fluence has not yet been determined for NMP-1. Since a capsule re-insertion program is in place at NMP-1, it will be possible to eventually develop a plant-specific model.

UTILITY PERSPECTIVE

The RG 1.99(2) model predictions showed that the USE for the critical beltline plate would drop below 68 J prior to EOL, but, data from the first two surveillance capsules indicated no shelf drop. The need to better understand the material-specific behavior of the reactor pressure vessel materials at Nine Mile Point Unit 1 became evident. In 1991, a research project was initiated to more accurately model what was occurring, taking into consideration material-specific data. This effort was part of a comprehensive research effort (which has been ongoing since the mid-1980's) to better address reactor vessel aging issues at Nine Mile Point Unit 1. In addition, this project was also performed in conjunction with a broader research effort sponsored by the Empire State Electric Energy Research Corporation in collaboration with Columbia University. As a result of these efforts, Niagara Mohawk Power Corporation (NMPC) now has a model capable of producing with high confidence RPV damage assessment. The model will be continually refined and verified as more data become available from the expanded surveillance capsule program.

SUMMARY AND CONCLUSIONS

Comparatively little work has been done in the past to develop a physically-based Charpy USE decrement model. Several plants have beltline welds and plates with USE levels near the 10 CFR 50, Appendix G 68 J screening criterion. The development of accurate and precise ΔUSE trend curves will increase in importance over the next decade as plants approach EOL. This paper presents the initial phase of work oriented toward identification of the variables which are important in terms of ductile fracture behavior of irradiated A302M steel.

Based on the preliminary work reported herein, it has been observed that a drop in the unirradiated USE requires an incubation dose. Therefore, the fact that the two NMP-1 surveillance capsules show no shelf drop is not surprising. The material-specific data seem to correlate well with square root of the dose, indicating the importance of the fine scale particles and the matrix damage component. The next datum will be of significance since it can be used to determine a plant-specific trend curve when used in conjunction with the material-specific slope.

ACKNOWLEDGEMENT

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