RADIATION EMBRITTLEMENT SATURATION EFFECT IN COMMERCIAL LIGHT WATER REACTORS

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SUMMARY

Recent surveillance capsules removed from commercial light water reactors have shown Charpy upper shelf energy and transition temperature shifts which are much less than would be predicted from test reactor data. The reason for the difference in these two data sets is postulated to be due to flux rate effects. The commercial light water surveillance specimens were irradiated in low fluence positions with lead factors of two or less and flux levels of approximately $3X10^{10}$ n/cm^2-sec. The test reactor data were developed with flux levels of a factor of two to ten greater than this value.

Preliminary modeling studies suggest that self-annealing phenomena are a major factor in this flux rate effect. An early model based on self-annealing accurately describes data from both high flux and low flux irradiations at temperatures less than 450^{o}F where diffusion rates are so low that self-annealing does not take place and at 550^{o}F where diffusion rates are high enough to cause self-annealing to be considered.

The present data base including the embrittlement saturation effect is too limited to propose regulatory changes at this point in time. An ongoing program is described involving surveillance capsules from ten commercial reactors which when completed should provide sufficient data to allow reevaluation of current radiation embrittlement prediction methods.

What is the saturation effect? Simply stated, in low flux irradiations, the normal manifestations of embrittlement (change in transition temperature, change in Charpy upper shelf energy, and change in flow properties) are affected only up to some level of exposure and any subsequent exposure does not affect these properties. This is in contrast to high flux irradiations where the manifestations of embrittlement are affected proportionally with fluence and no saturation is observed. The real significance of this observation results from the fact that the current embrittlement trend curves are based exclusively in the high fluence regimes (that is greater than $1x10^{19}n/cm^2$) on high flux irradiation experimental data. These irradiations are made either in test reactors or accelerated positions in power reactors. These trend curves are used to predict the state of embrittlement in reactor pressure vessels and consequently affect the heat-up and cool-down curves for such vessels and, in some cases, the operability of plants. The vessel materials are experiencing low flux irradiation, and therefore the damage should saturate early in life, as indicated in the low flux, high fluence irradiations. The materials are being penalized through the use of the trend curves that are based on high flux irradiations. Unfortunately, the existing data are too limited to build justify altering the present trend curves.

The significance of the saturation effect can be demonstrated by evaluating the current embrittlement issue. There are presently several U.S. reactor vessels currently under scrutiny by the U.S.NRC for possible embrittlement violations of 10 CFR 50 [1] and Regulatory Guide 1.99.1 [2]. Predictions made with the present trend curves of U.S.NRC Regulatory Guide 1.99.1 indicate that several of these reactor vessels will transgress the embrittlement limit of 50 ft-lbs [3] in the very near future. The same trend curves indicate continual degradation of the properties; consequently in the future, several of these reactor vessels may require a thermal anneal treatment to restore properties. If this saturation effect can be demonstrated and proved, most, if not all, of the reactor vessels in question can be vindicated. The probability of ever thermal annealing a reactor vessel can be minimized. In addition to the transgression of the embrittlement limit imposed on Charpy upper shelf energy, there is an operational problem associated with NDT shift. As discussed in the plant's technical specifications, the NDT shift determines the pressure-temperature limit curves for heat-up and cool-down. The predicted shifts for several PWR reactor vessels are in excess of 350°F at end-of-life fluence conditions, making operation difficult, if not impossible. The existing low fux data indicate that the trend curves can overestimate the NDT shift by as much as 60% to 75% at end-of-life conditions. Consequently, the severe operational restrictions imposed by the large NDT shifts can be reduced if the saturation effect can be proven.

A summary of the low flux, high fluence surveillance results is presented below. The data are developed from the surveillance programs of the Point Beach Unit 1 and the Connecticut Yankee nuclear power plant, which have seen 7.5 and 10 effective full-power years of operation, respectively. Figure presents the data from the Point Beach [4] weld metal which contains 0.24 wt% copper and 0.019 wt% phosphorus, at fluence levels ranging from 0 to $2.2x10^{19}n/cm^2$, it is important to note the change in the Charpy upper shelf energy, i.e., the maximum level obtained, as well as the shift in the transition of the Charpy energy as a function of fluence. The Charpy upper shelf does decrease initially with exposure up to a level of about $7x10^{18}n/cm^2$. At an exposure level 3 times that, there is no subsequent decrease in Charpy upper shelf energy. A similar effect is noticed with the shift in the Charpy curve. Note, for example, the 30 ft-lb energy temperature for the 3 capsules. This weld has an initial upper shelf energy of 65 ft-lb. This level of upper shelf energy, in conjunction with the copper level, makes Point Beach Unit 1 a possible candidate for a future thermal anneal if the current trend curve is used. These surveillance results indicate that at even end-of-life fluence conditions, the upper shelf Charpy energy is in excess of the 50 ft-lb embrittlement limit. Figure 1b. illustrates the saturation of the NDT shift. The two lower fluence capsules results indicated that the NDT shift was proportional to fluence to a level of $7x10^{18}n/cm^2$. The shift then saturated had a level of 210°F and remained constant to $2.2x10^{19}n/cm^2$. At this level, the trend curve predicts an NDT shift of approximately 330°. The next example of low flux irradiated materials is shown in Figure 2; this is an SA302 Grade B plate with a copper content of 0.19 wt% and a phosphorus content of 0.010 wt% [4]. The Charpy trend curves are shown in Figure 2a. and the Charpy shift summary is shown in 2b. Also indicated in Figure 2a. is the scatter band for the unirradiated Charpy data. All of the irradiated data lie within a similar band as shown. It appears from these results that irradiation may not increase the scatter in the Charpy impact energy. Again, the saturation in NDT shift is clearly indicated in Figure 2b. at a fluence level of $3.6x10^{18}n/cm^2$ or less. A comparison of results of the weld metal (Figure 1) and the plate (Figure 2) indicates that the level of saturation may be a function of not only chemistry but also microstructure. The results of the embrittlement of one of the Connecticut Yankee surveillance materials [4], an SA302 Grade B plate, is shown in Figure 3. Again, the saturation of embrittlement is clearly present.

All of the commercial reactor surveillance programs contain specimens machined from a reference material. For the older reactors, this reference material is an SA302 Grade B plate, with a copper content of 0.2 wt% and a phosphorus content of 0.011 wt%. A summary of the results from 7 surveillance capsule of the reference material is shown in Figure 4 [5]. The fluence levels range from a low of $2.1x10^{18}$ to a high of $2.2x10^{19}n/cm^2$. Figure 4a. illustrates the unirradiated Charpy curve as well as the data from the 7 capsules. Also indicated are the Charpy curves predicted by U.S.NRC Regulatory Guide 1.99.1 corresponding to fluence levels of $2.1x^{18}$ and $2.2x10^{19}n/cm^2$. The trend curves overestimate the drop in Charpy upper shelf energy and tend to underestimate the shift in NDT temperature at the low fluence level and overestimate the shift significantly at the high fluence level. This point is illustrated in Figure 4a. with the trend curve format.

Analyses of all the surveillance materials from the Point Beach Unit 1 plant and the Connecticut Yankee plant, as well as two materials from the R. E. Ginna plant [5] was performed to assess the difference between the predicted NDT shift and the actual NDT shift. The results of the analyses are presented in Figure 5. At low fluence levels, the predictions underestimate the actual shift in NDT. However, at fluences approaching end-of-life levels, the trend curve overpredicts the shift by as much as 80%.

It is obvious from the data presented that there is a significant difference between embrittlement experienced in the low flux irradiations typical of reactor vessel walls and high flux irradiations as in test reactors. One explanation for this difference is the occurrence of in situ annealing at operating reactor temperatures, normally 550°F. This annealing is the result of defect diffusion, and subsequent self-annihilation in the material. The defect diffusion rates in this material at 550°F are relatively low; consequently, the annealing process takes a significant amount of time. With the high flux irradiations, the time required to reach the desired fluence levels is too short for this annealing to occur.

Some preliminary modeling studies at the University of California, Santa Barbara, are evaluating this self-annealing phenomenon. The early results are shown in Figure 6 [6]. Analytical predictions for the shift in transition temperature as a function of fluence are made of low-temperature irradiations at both high and low flux levels. It appears that for irradiation temperatures less than 450°F, the diffusion rates are so low that the self-annealing effect is not present even for low flux irradiation. The model is extended to irradiations at 550°F, both for high and low flux rate. The self-annealing effect is obvious when the high flux and low flux predictions are compared in Figure 6 at fluence levels over $1x10^{19} n/cm^2$. Also included in the figure are appropriate data for the low temperature irradiations and the irradiations at 550°F for the two flux levels. The agreement between the low flux prediction at 550°F and the actual surveillance data is surprisingly good.

In order to alter the current trend curves contained in U.S.NRC Regulatory Guide 1.99.1 to account for this self-annealing effect present in low flux reactor vessel irradiations, more data are necessary. EPRI has tentatively identified 10 capsules from reactor surveillance programs that can be pulled and tested the next two years. Table 1 lists the plants from which these capsules will be removed.

Table 1

CAPSULES IDENTIFIED ON THE BASIS OF AVAILABILITY, AGE, FLUX, MATERIAL, AND OVERALL SURVEILLANCE PROGRAM

Plant	Utility	Approximate Fluence	Approximate Operation	Approximate Flux
		10^{19} n/cm^2	(FPY)	10^{10} n/cm^{2}s
Beaver Valley I	DPL	0.13	1.5	2.8
Zion I	CECo	0.75	4.0	6.0
Kewaunee	NSP	1.35	4.0	10.8
Point Beach I	WEP	1.48	5.0	9.4
San Onofre	SCE	4.60	8.0	18.2
H. B. Robinson	CPL	3.20	6.5	15.6
Conn. Yankee	YAP	1.60	8.8	5.7
Zorita	Spanish	8.5	6.5	41.5
R. E. Ginna	RGE	1.00	8.0	4.0
Big Rock	CP	1.60	11.5	4.5
Maine Yankee*	MYAP	0.24	5.0	6.4

*Alternative

As noted in Table 1, most of surveillance capsules are in the high fluence regime, i.e., greater than $.75 \times 10^{19} n/cm^2$, and the flux levels corresponds to lead factors of less than 3.0. There are two exceptions in the list. The first is a low fluence, low flux irradiation to be removed from Beaver Valley Unit 1. The purpose of this experiment is to assess the accuracy of the current trend curves in the low fluence regime. These data are to be compared with available Babcock and Wilcox low fluence surveillance embrittlement information as well as appropriate BWR surveillance information. Also included in the list of capsules is one from the Zorita plant from Spain. The fluence on this capsule is $8.5 \times 10^{19} n/cm^2$ and the flux level corresponds to a lead factor of approximately 9.0. All of the plants have Westinghouse NSSS Islands except for the Big Rock Point (General Electric BWR) and the Maine Yankee (Combustion Engineering PWR) plants. The data generated from these capsules are to be analyzed with the appropriate qualified surveillance data and appropriate trend curves are to be developed statistically to verify the existence of the irradiation embrittlement saturation effect.

REFERENCES

[1] "Licensing of Production and Utilization Facilities." Title 10, Chapter 1, Part 50, Federal Register, Vol. 38, No. 130 (July 1973).

[2] "Effect of Residual Elements on Predicted Radiation Damage to Reactor Vessel Materials," Regulatory Guide 1.99, U.S. Nuclear Regulatory Commission, Washington, D.C., July 1975, and Revision 1 (April 1977).

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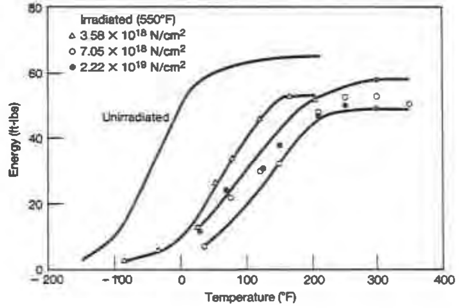
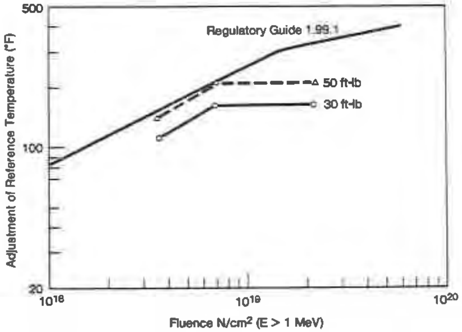
[4] YANICHKO, S., CHIRIZOS, J., "Assessment of the Validity of Trend Curves in Predicting Embrittlement of Reactor Pressure Vessels," Presented at the ASTM E-10 Symposium, Richland, Washington (June 1978).

[5] MARSTON, T. U., "The Embrittlement Saturation Effect Observed in Low Flux Surveillance Irradiations," Technical Summary Report, Electric Power Research Institute (December 1978).

[6] WULLAERT, R. A., et al., "Analysis of Radiation Embrittlement Reference Toughness Curves," EPRI RP886-1 Semiannual Progress Report No. 4, March 11, 1978 - September 10, 1978, Fracture Control Corporation (March 14, 1978).

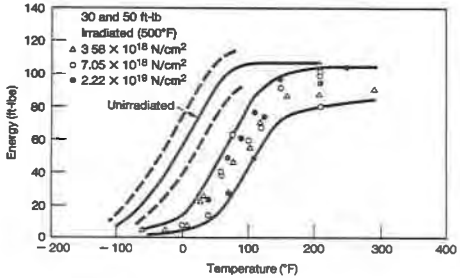


a Charpy data for three fluences.

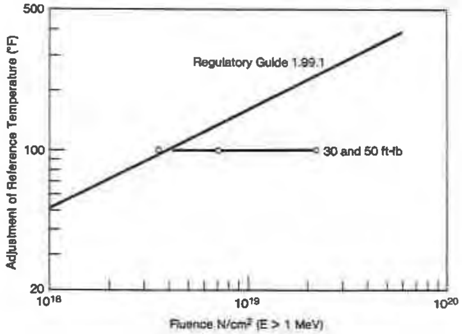


b Actual shifts compared with Regulatory Guide 1.99 1 predictions; note apparent saturation at 8 × 10^{18} nvt.

Figure 1. Low Flux Irradiation Results for Point Beach Unit 1 Weld Metal (0.24 weight percent Cu, 0.019 weight percent P)

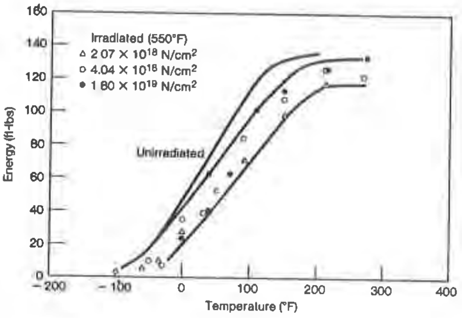


a. Charpy data; note unirradiated scatter band relative to irradiated data.

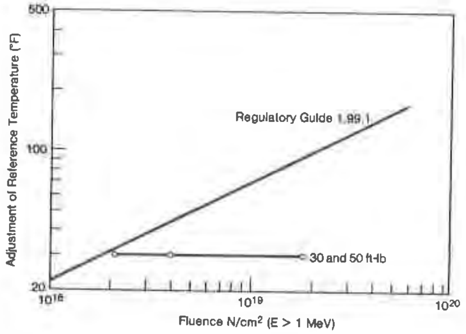


b Actual shifts compared with Regulatory Guide 1.99 1 predictions; note apparent saturation at 3 × 10^{18} nvt or below.

Figure 2. Low Flux Irradiation Results for Point Beach Unit 1 Plate (0 19 weight percent Cu, 0.010 weight percent P)

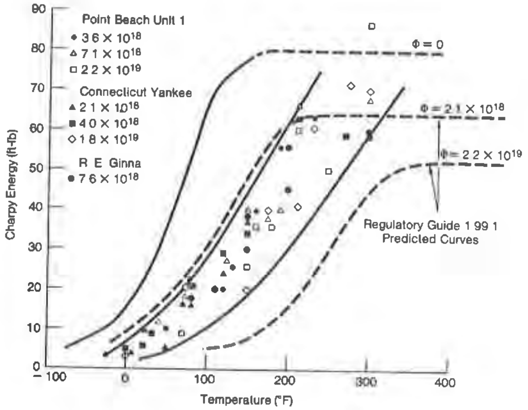


a Charpy data for three fluences



b Actual shifts compared with Regulatory Guide 1 99 1 predictions; note apparent saturation at 2×10^{18} nvt or below.

Figure 3. Low Flux Irradiation Results for Connecticut Yankee Plate (0 10 weight percent Cu, 0.010 weight percent P)



a Charpy data for seven fluence levels with predicted Charpy curves (Regulatory Guide 1.99.1) for the lowest and highest levels.

Figure 4. Low Flux Irradiation Results for ASTM Reference A 302 B (0 20 weight percent Cu, 0.011 weight percent P) From Three Reactors

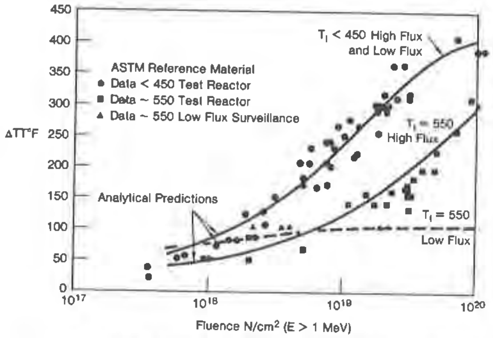


Figure 6. Preliminary predictions of ΔTT shift for low temperature (T_i <450°F) - high and low flux irradiations; for operation temperature (550°F) and high flux test reactor irradiation and operating temperature (550°F) and low flux, surveillance irradiations. Also included are available data for the three types of irradiations.

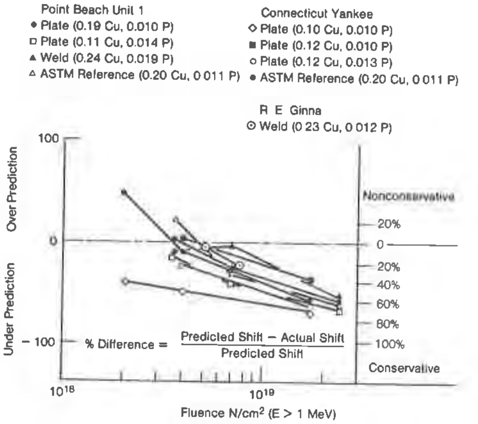


Figure 5. Summary of the error in prediction of the ΔTT shift for eight materials irradiated in reactor surveillance (not accelerated) capsules. Note as the fluence approaches end-of-life conditions, the Regulatory Guide 1.99.1 over predicts the shift by approximately 60 to 70 percent.