Review of

Technical Basis for Revision of Regulatory Guide 1.99: NRC Guidance on Methods to Estimate the Effects of Radiation Embrittlement on the Charpy V-Notch Impact Toughness of reactor Vessel Materials

NRC Reg. Guide 1.99 Revision3

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December 2007

This report contains a review of the NRC Regulatory Guide 1.99 Revision 3. As requested in the Statement of Work, the review first addresses the soundness of the technical bases supporting the recommended revisions to the Reg. Guide. Other major issues that bear on the technical accuracy of the report are discussed next. Less critical issues are treated separately. While their resolution does not bear on the technical accuracy, they can affect the clarity of the report. Finally, minor errors in the text are noted in the last section.

Overall, the technical basis for the recommended revisions in NRCs Reg. Guide 1.99 R3 are adequate and are consistent with the current technical state of knowledge published in the open literature.

SOUNDNESS OF THE TECHNICAL BASES SUPPORTING THE MAJOR RECOMMENDED REVISIONS TO NRC'S REG. GUIDE 1.99 R3.

1. ΔT_{30} Embrittlement Trend Curve

The technical bases for the recommended ΔT_{30} embrittlement trend curve in NRC's Reg. Guide 1.99 R3 are generally adequate and are, for the most part, consistent with the current technical state of knowledge published in the open literature. There are several points that should be addressed in solidifying the soundness of the trend curve. These are discussed in the subsequent subsections.

1.1 Flux effect

A flux effect is given in the MD term but not in the CRP term for the trend curve. In particular, Figures 4-13 and 4-14 show the effect of flux on saturation fluence and on the parameter, B. These data come from the IVAR database, which was built to uncover a flux effect. As stated on p. 4-2, one of the purposes of the IVAR database is to motivate the functional form of the candidate fitting function. This is done in Appendix A that deals with the flux effect on CRP hardening, Appendix A, section 2.3. Therefore, it is surprising that this effect is not incorporated into the model for ΔT_{30}.

The December 4, 2007 memo by Mark EricksonKirk provides a revision to the part of the Φ_{SAT} expression in the CRP term that incorporates effects of flux and nickel content. This revision is well supported by both the current understanding of how flux and nickel content should affect CRP hardening, and data from the IVAR and RADAMO databases. ***It is recommended that this model be considered for inclusion in NUREG 1.99 rev. 3.***

1.2 Database selection and logic

The logic for choosing the database(s) on which to develop and validate the models is not clearly explained a priori. Database selection and use is detailed in Section 4.2 of the report and afterward. As stated there, the entire US surveillance database is used to fit the embrittlement trend curve and to fit the upper shelf energy. The components of the ETC (MD and CRP) are first fit separately and then the fits were combined and assessed on the same database used for the separate fits. The goodness-of-fit was determined by minimizing the T-statistic on slopes and intercepts of the equation:

$\Delta T_{30(Predicted)}\ \Delta T_{30(Measured)} = m\Theta + b.$

The best fits were then assessed for their predictive ability on data that were not used in developing the fits. The assessment was used to modify the best-fit equation to apply to a larger range of conditions.

While this logic is contained in the report, the databases are discussed before their roles in the report are explained, leading to confusion. As early as sub-section 4.2, the IVAR database is mentioned. Therefore, ***the recommendation is to insert a sub-section at the beginning of Section 4 that describes the various databases and how they will be used in this report.*** Then any reference to the various databases can be understood in the context of its role in the report.

1.3 Use of databases in the calibration of the ETC

There are several issues that should be addressed concerning the use of databases in the calibration of the ETC. These are presented in the following sub-sections

A. What is the logic for using the databases as described in this report? For example, why was the entire US surveillance database used to fit the models? A more standard approach is to use a fraction to fit and the remainder to assess or verify. Using the entire database to fit the models left nothing to verify them on a database of the same nature. Alternatively, one could make several small random samples (with each sample consisting of 10% of the data) to fit the data and then calibrate the model on the remaining 90% of the data. This 10-fold sampling actually uses all of the data for fitting and all of the data for validation, but provides a means of validating the fits with the same dataset used to calibrate.

B. The report adopted the position that model fitting could only be done with the US Surveillance database. However, confirmation was then done using other databases. What is the wisdom of this approach vs. fitting and validation within a single database? Some discussion of the justification of the process used in the report should be provided.

C. Another drawback in using only the US Surveillance database for fitting is that it is unable to properly assess several effects such as the Ni effect in the MD term, the Mn effect in the CRP term and flux effect above 4×10^{10} n/cm^2s. These dependencies could be fit using the IVAR database, for example, and then calibrated within the overall US surveillance database to at least check for inconsistencies. In this way, dependencies could be incorporated into the models.

D. What is the drawback in crossing databases to assess the capability for extrapolation? This logic was followed in the report, but it necessarily involves jumping to a different database, which could dictate different fitting parameters *within* the parameter space that the functions were originally fit.

E. Why was fitting limited to the US surveillance database? One could argue that it is the most complete and carefully constructed database. However, it is clearly not complete enough to capture several parameter effects. If there is doubt about the quality or legitimacy of the other databases, then one could argue that they should not be used at all in the construction of the final models. In that regard, excluding the IVAR database is particularly puzzling as this database was constructed under

NRC funding and scrutiny and involves parameter identification that was simply not possible in the US surveillance database.

Mark EricksonKirk has provided additional information (not contained in the report) on the lack of success in calibrating a fit to a combination of databases such as the US-LWR, IVAR and RADAMO databases. It is recommended that this discussion be included in the report to justify the confinement of fits to only the US-LWR database. It would also be beneficial for the report to contain an assessment of the advantages and the drawbacks of using test reactor databases in the calibration process.

As a final note, ***it is recommended that test reactor data be utilized to the fullest possible extent in fitting the ETC at high fluence***. The US-LWR database is clearly lacking in high fluence data. Yet the importance of high fluence data is critical to the prediction of behavior beyond the 40-year license period. Test reactor databases extend to higher fluence and thus these databases should be used to inform the ETC as to the form of the fit at higher fluence. While the report cites the drawbacks to the use of this data, this reviewer believes that the benefits outweigh the drawbacks.

2. <u>ΔUSE Equation</u>

The functional relationship between ΔUSE and ΔT_{30} is: $\Delta USE = 1.18 \bullet \Delta T_{30}$, where ΔT_{30} is in °F and ΔUSE is in ft-lbs. This linear relationship is expected based on the available data, and it is also be justified based on the commonality of the physical processes governing the ductile-to-brittle transition temperature and the upper shelf energy. In particular, the TUS FFEMS paper by EricksonKirk and EricksonKirk makes a compelling argument for a physical basis that supports the correlation between ΔUSE and ΔT_{30} data. ***This data and the description of the physical basis supporting the relationship should be included in Revision 3***. The only concern regarding the analysis in the paper is the assertion that temperature dependence is controlled only by the lattice structure and not metallurgical parameters such as precipitates, grain boundaries, etc. That statement implies that temperature cannot affect the processes governing the interaction between dislocations and the particles responsible for hardening. However, thermal energy can induce climb of dislocations over obstacles and it can aid in the cutting of the particles. In this way, metallurgical parameters do affect the temperature dependence of deformation/hardening.

3. <u>Attenuation Equation</u>

The discussion regarding the attenuation term is a bit ambiguous and misleading, or perhaps, incorrect. This term provides the flux as a function of vessel wall thickness given the flux value at the ID and the depth into the vessel wall. It is stated on p. 8-1 that "Eq. 8-1 conservatively assumes that fluence attenuates like displacement per atom (dpa) (i.e., Eq. 2-7 assumes that fluence attenuates more slowly than it actually does)." This statement cites Randall 87 as its source.

However, Randall's paper (p. 162) states that "to convert to a 'dpa equivalent' formula, we used some calculations reported at the 4th ASTM Euratom Symposium [8], which showed that the dpa attenuation through an 8.0 in. vessel wall is less than the attenuation of fluence, n/cm^2 (E>1 MeV) by a factor of 2.06, the average of six calculations made for different reactor vessels." Thus, the relation in Eq. 2-7 (or 8-1) is in units of fluence but follows the dpa behavior. This is why the exponent is -0.24 vs. -0.33. Hence, the conservatism of the dpa behavior is already incorporated into Eq. 8-1. So statements to this effect are incorrect.

Further, the conclusion from MRP-56 is curious. Why should it be included in this section or in the report at all? It provides no support for the form of the attenuation term and in fact, argues against the use of fluence-based modeling.

OTHER MAJOR ISSUES

4. DPA- vs. Fluence-Based Hardening Models

The issue of fluence vs. dpa was raised in section 8 of the Reg Guide. While it is the intent of the Reg Guide to use physically based models, it could be argued that the use of fluence rather than dpa contradicts this intent. In fact, dpa is a much better descriptor of neutron damage leading to changes in hardening and embrittlement than is fluence and will therefore result in much better physically based models. It is curious that this point is not addressed anywhere in this revision. My understanding is that for surveillance specimens that are located at the ID of the RPV, there is no significant attenuation in any of the coupons and that over the range of power reactors used in the surveillance database, the fluence and dpa can adequately be related by a constant. However, that point is not discussed anywhere in this report. ***It is therefore recommended that a section be added that addresses the relation between fluence and dpa, and that***:

- the issue of dpa-based damage be noted as the best way to express the effect of neutrons on structural materials properties,
- a section be added containing a discussion of the relation between fluence and dpa for the databases used in this report and that the case be made and supported for why fluence is an adequate representation of neutron damage in RPV steels.

While it is, perhaps, too ambitious for Revision 3, ***it is also recommended that plans be made to address the issue of dpa-based models of temperature shift in the next revision***. What is required is the development of a relation between fluence and dpa, followed by conversion of the database from one based on fluence to one based on dpa. Then the models will need to be reformulated to capture the different dependence of the various factors on dpa vs. fluence.

5. Functional Form of Fluence Dependence of Hardening

The under-prediction of hardening at high fluence is noted on p. 4-65. An equally important issue is whether the functional dependence on fluence ($\Phi^{1/2}$) is correct. By its form, a $\Phi^{1/2}$ functional dependence implies infinitely increasing damage with fluence. This is not physically reasonable as the damage will saturate at some value of fluence. While the saturation value may not lie within the fluence range of the database, it may become important at fluence values in the 10^{20} n/cm^2 range that will be important for licensing extensions to 60 and 80 years.

6. <u>Embrittlement Mechanism at High Fluence (> 3 x 10^{19} n/cm^2)</u>

It is clear that with the prospect of life extension to 60 and perhaps 80 years and even beyond, a model that can accurately predict embrittlement trends beyond 1 x 10^{20} n/cm^2 is critical to the viability of these extensions. Unfortunately, this is precisely the fluence space where the surveillance data is most sparse. Test reactor irradiations provide some data in this regime, but their use also has drawbacks. Yet a more significant issue is the lack of understanding of the embrittling process in this fluence range. (If the process was understood, then physically based models could be employed to capture the proper dependencies and perhaps, reduce the amount of data required to calibrate them.)

Above a fluence of 3 x 10^{19} n/cm^2, CRP hardening has saturated and MD hardening is increasing, perhaps rapidly. On the basis of test reactor data, it could be deduced that the difference between measured and predicted values is due to an embrittling process that we do not yet understand, and that we have not yet characterized. For these reasons, it will be imperative that existing samples from high fluence irradiations are characterized and that an effort be made to understand the embrittling process in this fluence range. Because it will be difficult to "catch up" to the required fluence levels in time to inform the license extension process, the only avenue may be a more highly accelerated irradiation program than has been conducted to date, and that perhaps relies on the use of ion irradiation.

LESS CRITICAL ISSUES

1. MD and CRP hardening regimes are separated by the Cu content of the steel. Some documentation establishing the regimes of dominance and justification of the transition Cu concentration below which MD controls hardening and above which CRP contributes, would be helpful. For example, Figures 4.1/4.2 and Figures 5.3/5.4.

2. p. 4-82 – The gross failure of RM-(2) in fitting to the JNES test reactor database. The error is astronomical. If this is an issue of poor fits at high fluence, then does it imply that there are significantly different processes occurring at higher fluence than occur at low fluence? If so, then this is a very important issue to address.

3. pp. 9-4 and 9-5 – there is an inconsistency in how the cut-off between low and high fluence is defined. Section 9.4.1 (p. 9-4) gives the ΔT_{30} trend for fluence at or below 2×10^{19} n/cm^2. On p. 9-5, point (1) in the second paragraph states that the temperature shift model is applicable for $\Phi < 3 \times 10^{19}$ n/cm$^{2.}$ This cut-off value of fluence doesn't agree with that given on the previous page.

MINOR PROBLEMS WITH THE TEXT

These comments focus on mainly minor issues in the text, such as typos, missing labels or data, unclear or unsubstantiated statements or arguments and problems with the logic.

Comments on this and the next page were made on the August 6, 2007 version of the report.

p. xvi - insert comma between fluence and YS symbols - two places.

p. xvii - units should be consistent. Why is DT30 in units of degrees F while the T in the eqn for DYS(MD) is in units of Kelvin?

p. xviii - Is this eqn 4-18? If so, then it needs to be identified.

p. xx - "2" should be superscript in the unit of fluence.

p. xxi - remove either "is" or "cannot" in first line of section on "Flux".
- Change "property" to "properly" in 6th line in section on "Flux".
- Third line from bottom of "Flux" section - update reference to correct section in the text.

p. 1-1 - Change "the" to "then" in first line below bullets.

p. 1-2, 10th line from the bottom – Eliminate "be" after "should."

p. 1-3 line 1: Correct "informed the information"

p. 2-3, part b.i. - Are "sufficiently small" and "unambiguously" quantified?

p. 2-4 - units for fluence should be consistent throughout. Either use capital "phi" or lower case "phi" x t.

p. 3-3: Footnote below Fig. 3.3 Something is missing in the second to the last line.

p. 4-1, line 1: insert "of" after "development".

p. 4-2, line 4: change "sauce" to "such".

p. 4-2 line 6: change "un-observed" to "not observed".

p. 4-2, line 2 of pph 2: eliminate "/".

p. 4-2, definition of saturation fluence. Remove "of the percent".

p. 4-2, section 4.1.1, line 1: should be "the point defects" not "point the defects".

p. 4-2, section 4.1.1. Vacancy clusters are not really nano-voids. The clusters are closer to a high density of vacancies in the lattice. Suggest dropping "nano-voids". Last sentence is not strictly correct. Solutes acting along can do so as "atmospheres." They can act as second phases but they must form a distinct phase, generally with another element in the matrix. So they can't act as "second phases alone". They can also affect hardening by binding to vacancies, called solute-vacancy clustering.

p. 4-3, Fig. 4-1 - Caption should read: "Data showing that within the fluence range of relevance to LWR PRV steels, the yield strength is proportional to the $(\text{fluence})^{1/2}$. The proportionality breaks down at doses well above the upper limit of the fluence range.

p. 4-3, sec. 4.1.1.2, line 3: Change to "...more prone to increased recombination and loss to sinks."

p. 4-10, Fig. 4-2 caption. Suggest eliminating embedded figure numbers and captions.

<u>Comments from this point forward are based on the October 1, 2007 version of the report</u>

Figs 4-3, 4-5 and 4-7: The max Si wt% is incorrect. What is the proper value?

Fig 4-4 - remove embedded figure caption.

Fig 4-5, 4-4: It would be more useful if the units used to describe the hardening are consistent between
these figures.

p. 4-12: This footnote appears to be incomplete.

Fig. 4-6: eliminate embedded caption.

p. 4-14: This statement is predicated on the fact that MD hardening can be separated from CRP hardening. This should be established.

p. 4-14: It would be valuable to show the tanh fit to substantiate the point made here.

Fig 4-8. It is not clear that there is no effect of Cu on hardening. At least the effect shown is no less than the flux effect shown in Fig 4.3, which is considered to be real.

p. 4-17, pph 1. I have a lot of problems with the comment that the correlation between the Chaouadi curve and the IVAR database is "good." First, on what is the judgment based? There should be some sort of quantitative characterization of the fit before a judgment is proffered. Second, what is the "best fit" to the data and how does that compare to the Chaouadi curve? Lastly, if one removes the curve, the data looks to contain a large amount of scatter that will be difficult to fit "well" with any curve. Without a priori knowledge of the Chaouadi curve, I doubt I would have drawn a best fit that resembled it.

p. 4-17: In discussing CRP hardening, as with MD hardening, the report needs to explain how these components of hardening are broken out separately for evaluation. That is, how is the portion of hardening due to CRP known, or alternatively, how is the MD contribution subtracted from the total to arrive at the CRP fraction?

p. 4-18: What does the "high", "medium," and "low" refer to in Figs. 4-11 and 4-12. Caption and text need to reflect this.

p. 4-18: Is it assumed that the solubility limit is not affected by irradiation? Has this been verified for, by example, determining the amount of Cu in CRPs and subtracting from the total to verify the solubility of Cu in ferrite?

p. 4-19. Insert "excess" before "copper" in line 1 of pph 1.

bullets on p. 4-19. These "excess" Cu levels are a small (10-30%) fraction of the Cu in solution. So the issue of the effect of irradiation on the solubility of Cu is even more important. Also, given that only 0.03 Cu is needed for CRP formation to occur, knowing the amount of Cu in second phases is also important. Are these second phases stable under irradiation?

p. 2-4: The f term on the RHS of eqn 2.7 should have a subscript “ID”.

p. 2-4, last pph: Change to “see Appendix A to Appendix C”.

p. 4-19, sec 4.1.2.2 - looks more like 25% to me. But there is not a lot of data to support a hard quantitative value.

p. 4-19. Eliminate first "for" in line 10 of section 4.1.2.3.

p. 4-19: Sentence on the integrated effect of flux-induced changes is misleading. It implies that the saturation flux increases with decreasing flux.

p. 4-20: sec. 4.1.2.4 - the data suggest to me that P has a comparable effect on both B and Φ_{sat} as does Ni. Why was it discounted?

p. 4-24, section 4.1.3. So shouldn't hardening from different sources be determined in a way that is sensitive to their relative contribution? That is - not just one rule or another, but perhaps a mixture?

p. 4-26: Change E_{temp} in line 2 to E_{flux}.

Table 4.2 - entry in box under CRP in Fluence row - entry is unclear: B 1.5.Phi_SAT. What is this?

p. 4-29 - first bullet. Is it that impurity segregation "will" not occur or that it will not occur to the extent needed to have a measurable effect? I would think it is the latter.

p. 4-30: Should read "Eq. 4-4".

p. 4-32. The fitting process focused on 2 variables, T_TOTAL and T_MAX. But what is the criteria used to determine the "best" fit, min T_MAX or min T_TOTAL?

p. 4-38, Fig. 4-24 caption. Should read "IVAR" not "PVAR".

p. 4-42, line 1. Change "MD" to "CRP".

p. 4-42, bullet #2. Change "*" to "-".

Fig. 4-28: Define FAST if it has not already been done.

eq. 4-10, p. 4-55. Note that beta has units of °F/MPa.

p. 4-60, item (b) line 4: change "this" to "the".

p. 4-61, bullet (c), line 3: Change to "the slight increase in saturation fluence in flux..."

Fig 4-32. Call out "BR2" in the legend in the graphs to be consistent with the text.

p. 4-64, second line below table. Why are D and E determined by eye and not statistically?

p. 4-65 line 2 in sec. 4.3.2: Change "Eq. 4-3" to "Eq. 4.4".

p. 4-65: Eq 4-14 is not the same as Eq. 4-4 and the argument about the two terms affecting the peak CRP hardening is unclear.

Fig captions 4-38 and 4-39. Remove "both" before 290°C.

Fig 4-42. The agreement is horrible - almost an anti-correlation - 200% error?

Fig. 4-47, 4-49: Change "D" in x-axis labels to "Δ - Greek symbol".

p. 5-1, 3rd pph, line 1: Something missing after "from".

p. 5-1. Reference statement in last line in pph 3.

Fig 6-1 legend. Should read "...in Appendix A to Appendix C."

p. 8-1 first line of pph 2. Insert "a" before "review".

p. 8-1: lines below the equation are confusing. The equation is variously referred to as Eq. 2-7 and 8-1 in the same sentence when in fact these are the same equation.

p. 9-6: Section 9.4.1.3 Heading should read "…Between 2 x 10^{19} n/cm^2 and …."