LWR Pressure Vessel Embrittlement and Conditions of Validity of Current Predictive Methodologies

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Prepared For: 22.314

Prepared On: December 7th, 2006

Abstract

Because of the irradiation coming from the core, the beltline region of LWR vessels experiences a slow degradation of its mechanical properties, which is known as Reactor Pressure Vessel (RPV) embrittlement. An accurate prediction of the severity of this phenomenon, at each time during the operating life of the RPV, is fundamental in order to prevent the vessel from experiencing stress states that may lead to a non-ductile propagation of a critical crack that may be present in the vessel walls, thus yielding vessel failure.

Most of the literature, as well as current NRC regulations, relate the severity of embrittlement to fast neutron fluence only, neglecting the irradiation time rate as well as the effect of gamma-rays. In some circumstances, however, this procedure leads to an underestimation of the severity of embrittlement. Irradiation rate plays an important role when RPV walls experience a low fast neutron flux, like in BWRs. Gamma-rays give a significant contribution to embrittlement when a thick water gap separates the core from RPV walls, like in the ABWR. The current design tendency of decreasing fast neutron flux at the RPV wall, coupled with the introduction of new and innovative reactor features, is making the mentioned circumstances more and more frequent, requiring a revision of the current fast-fluence-based regulations.

Scope of the paper

The present paper is focused on RPV embrittlement in LWRs. It is aimed at providing practical information concerning (in the order they are presented):

- the microscopic causes of embrittlement;

- the macroscopic effects of embrittlement and its indicator parameters;

- the methodologies used to quantitatively estimate the indicator parameters;

- the process used to recover the mechanical properties degradated by an excessive level of embrittlement;

- the regulations aimed at preventing severe consequences that may be made possible by an excessive level of embrittlement;

- the limitations of the current regulations and their proven inadequacy in some circumstances.

The paper has a practical nature, in that it was written with the purpose of highlighting ready-to-use information and "rules of thumb", whose importance is often underrated.

1. Introduction

With respect to the vast majority of the mechanical components used throughout the industry, those aimed at in-vessel nuclear applications must be provided with an additional feature consisting of a "satisfactory behavior" under the effect of nuclear irradiation. This "satisfactory behavior" essentially consists of preserving, at some extent, the pre-irradiation characteristics, such as mechanical properties and geometry. The component representing the boundary between the high and the low radiation regions, i.e. the RPV, does not constitute an exception and it must satisfy this requirement too. In spite of absorbing a lower dose per unit time than most of the internals, the RPV is a non-replaceable component which is designed for a 40-60 year life. The cumulative damage experienced in this long time interval causes a slow modification of the mechanical properties of the RPV in the beltline region, which mainly materializes in the form of toughness reduction (embrittlement) and increase of the Ductile Brittle Transition Temperature (DBTT).

2. Microscopic causes of RPV embrittlement

The microscopic causes of embrittlement lie in the forming of obstacles to dislocation motion, called "hardening centers", as well as to changes of the composition and structure of the microscopic interfacial regions along which crystal plane sliding occurs. Both these phenomena are caused by several types of radiation-matter interactions, most of which are, in LWRs, induced by fast neutrons. Also γ-rays, in less frequent circumstances, may give a substantial contribution. From this consideration it follows that fast neutron irradiation is not the only cause of vessel wall embrittlement, even though its contribution is very often the dominant one.

The following subsections describe briefly the mechanisms causing embrittlement.

2.1 Neutron irradiation

Neutrons cause embrittlement mainly through two mechanisms: atom displacement and induced Helium production reaction.

The microscopic structural damage resulting from the collision between an incoming neutron and a stationary nucleus is more extensive when the incoming particle is highly energetic, i.e. when the neutron is a fast neutron. However, an accurate prediction of the radiation damage can not account only for the consequences of the occurrence of a postulated collision event, but also for the probability that this event can actually occur. In this regards, thermal neutrons turn out to have a moderately high displacement cross section. Figure 1 shows that, as the neutron energy is reduced, the atom displacement cross section, after going through a minimum of about 0.1 b near 1 keV, increases reaching about 18 b at 0.01 eV, a value which is only one order of magnitude lower than that characterizing 1 MeV neutrons.

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Based on Figure 6, Mansur and Farrell ([1]) state that "*thermal neutron induced displacements become significant (≥10%) with respect to those from fast neutrons*" when:

$$\frac{\Phi_{nt}}{\Phi_{nf}} \geq 10 \qquad (1)$$

where Φ_{nt} and Φ_{nf} are the thermal and fast neutron flux respectively. This situation is however quite unlikely since, even in the core of LWRs, the thermal flux is lower or comparable to the fast flux.

Neutrons cause vessel embrittlement also by inducing the ^{10}B(n, α)^{7}Li reaction, which has a larger cross section for thermal neutrons than for fast neutrons:

$$n + {}^{10}B \rightarrow {}^4He + {}^7Li$$
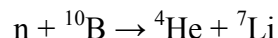
Since boron is often present in RPV steels as impurity, neutron irradiation can yield helium production. Then, the bubbles so formed tend to coalesce at the grain boundaries, initiating the so-called grain-boundary cracking, which causes structure embrittlement.

2.2 Gamma-ray irradiation

Several are the reactions that give origin to γ-rays. A strong source is the fissioning fuel in the reactor core, but also the peripheral structure materials can interact with thermal and fast neutrons and undergo reactions in which such radiations are emitted. Then, they may cause atom displacements in the vessel wall material by transferring their energy to electrons via Compton scattering, pair production and photoelectric effect. Figure 2 shows however that the atom displacement cross section for γ-rays, in the energy region of interest for nuclear reactors, is more than one order of magnitude lower than that characterizing thermal and fast neutrons.

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Again, by using the cross section as comparison tool, Mansur and Farrell ([1]) conclude that in order that γ-induced displacements contribute more than 10% compared to fast neutron induced displacements the following condition must be satisfied:

$$\frac{\Phi_\gamma}{\Phi_{nf}} \geq 100 \qquad (2)$$

where Φ_γ is the γ-ray flux. As discussed in Section 7.2, this inequality, not satisfied at the RPV walls of LWRs, can become true in case of wide water gaps interposed between core and vessel walls. This fact significantly affects the applicability of the current embrittlement-related regulations, which are "neutron-fluence based", to reactors having the mentioned characteristic. The ABWR is an example of such reactors.

3. Embrittlement: macroscopic effects, indicator parameters and quantitative prediction

3.1 Macroscopic effects of embrittlement

As mentioned before, embrittlement is the reduction of toughness of a material, which is in turn the maximum energy that the material can absorb before rupturing. Being this energy proportional to the area underneath the stress-strain curve, Figure 3 shows how significant this effect can be for a very embrittlement-sensitive metal, i.e. copper, when it

is irradiated to different neutron fluence levels. A secondary consequence of irradiation, i.e. the increase of yield and ultimate strength, is also evident from Figure 3.

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3.2 Indicator parameters for embrittlement
The most well known indicator parameter for embrittlement is not the toughness reduction itself (whose value is however accounted for and its estimate is discussed at the end of Section 3.3), but the reduction of the temperature at which the brittle-ductile transition occurs. In fact, toughness also depends on the temperature of the material of interest and, in general, the lower is the temperature the more brittle is the material[1]. The transition from brittle to ductile behavior, as a function of temperature, appears in the form of two plateaus separated by a narrow transition region, as shown in Figure 4. The toughness corresponding to the ductile behavior is called Upper Shelf Energy (USE), while that corresponding to the brittle behavior is called Lower Shelf Energy (LSE). Irradiation reduces USE and shifts the transition region to the right, thus increasing the temperature at which the brittle-ductile transition occurs. This temperature is usually referred as Nil Ductile Temperature (NDT), Ductile Brittle Transition Temperature (DBTT), or Reference Temperature for Nil Ductile Transition (RT_{NDT}).

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[1] More precisely, once transition from brittle to ductile behavior has occurred, most materials experience a very slow toughness reduction as the temperature is further increased.

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Since the brittle-ductile transition does not occur at a single temperature, but over a temperature interval, Appendix G of 10 CFR Part 50 ([4]) recommends to refer the increase of the temperature transition from the unirradiated condition, ΔDBTT, to the measurement performed at an impact energy of 30 ft-lb (41 J), as shown in Figure 4.

3.3 Quantitative prediction of embrittlement

Should experimental data not be available, Section 50.61 of 10 CFR Part 50 ([4]) recommends, with some limitations discussed later, the use of the following relation for ΔDBTT (ºC):

$$\Delta DBTT = \frac{5}{9} \cdot \left[CF \cdot f^{(0.28-0.10\log_{10} f)} + M \right]$$

(3)

where:

- f is the fast neutron fluence, in units of 10^{19} n/cm^2;

- CF (ºF) is a "Chemistry Factor", dependent on Cu and Ni content. Because of the embrittlement-enhancement effect caused by these two elements, CF increases as the Cu and Ni content increases, ranging from 20ºF (0 wt-% Cu, 0 wt-% Ni) to 320ºF (0.4 wt-% Cu, 1.20 wt-% Ni)[2];

- M (ºF) is a coefficient added to obtain conservative, upper bound values of the post-irradiation DBTT. It accounts for uncertainties in the analytical relations as well as in the knowledge of the pre-irradiation DBTT. A typical value for M is 56ºF for welds and 34ºF for base metal ([3]).

As mentioned above, equation (3) is not universally applicable, and its use is recommended as long as ([5]):

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[2] Chemistry Factors for several Cu-Ni content combinations are listed in Table 1 and Table 2 of Section 50.61 of 10 CFR Part 50 ([4]).

- the material under investigation is a ferritic steel of type SA-302, 336, 533, 508 (typical RPV steels);

- the minimum specified yield strength, in unirradiated conditions, is 345 MPa;

- the irradiation took place at a nominal temperature of 550ºF (288ºC).

Figure 5 shows the DBTT increase obtained by applying equation (3) to two RPV steels: a modified SA302B type (0.23 wt-% Cu, 0.49 wt-% Ni), and a low Cu/P-content steel, i.e. SA533B-1 (0.07 wt-% Cu, 0.60 wt-% Ni) (compositions from [3]). It is evident how significantly alloy elements can affect the increase of DBTT[3]. Figure 5 also shows the asymptotic nature of the irradiation-induced embrittlement: the rate of DBTT increase tends to decrease as the neutron fluence increases.
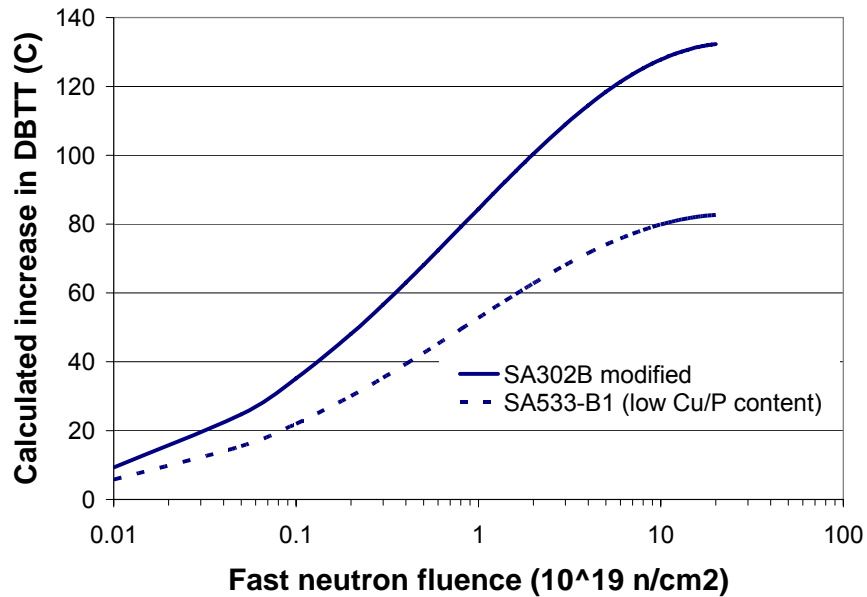

Figure 5: DBTT increase as a function of fast neutron fluence (E_n>1 MeV) for two types of ferritic steel. Curves obtained from equation (3).

The user of equation (3) must be aware of the significant dependence of the post-irradiation DBTT on the temperature at which irradiation takes place. Regulatory Guide 1.99, Rev.2, ([5]), states that, with respect to the values predicted by equation (3): "*irradiation below 525ºF (274ºC) should be considered to produce greater embrittlement, and irradiation above 590ºF (310ºC) may be considered to produce less embrittlement*". Figure 6 shows that irradiation experienced below 400ºF (204ºC) yields a post-irradiation DBTT that can be up to 100ºC higher than that resulting from an irradiation at 550ºF (288ºC).

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[3] It is important to point out, however, that Cu and Ni content can not be indefinitely reduced since the corrosion-resistance properties of the steel would be negatively affected.

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If together with the DBTT increase, the actual toughness reduction was also of interest, Regulatory Guide 1.99, Rev.2, ([5]), provides an useful graph to conservatively estimate the reduction percentage in Upper Shelf Energy (USE) as a function of neutron fast fluence and Cu content. This graph is represented in Figure 7 together with the fluence ranges for typical BWRs and PWRs.

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4. Toughness recovery process: thermal annealing

The loss of toughness experienced by the RPV beltline region can be partially (and, in some cases, totally) recovered by means of the thermal treatment typically referred as thermal annealing. Regulatory Guide 1.162 states that thermal annealing is "*the only known method for restoring toughness properties to materials degraded by neutron radiation*" ([7]). It consists of heating the RPV walls to a higher temperature than the operating temperature, for a sufficient period of time. This process can be performed in two ways:

- wet annealing: the annealing temperature is typically below 345ºC since water is used as heating medium. Although the relatively low temperature prevents achieving a complete toughness recovery, the process is not complicated from an engineering viewpoint since the primary water temperature is controlled by pump heat up, and 345ºC is a typical value for the RPV design temperature, at which the RPV is tested during preservice testing.

- dry annealing: the heat treatment is performed in air, by interposing a radiant heating source between core and RPV walls. Even though this technique allows reaching temperatures up to 430-480ºC, and therefore a significant toughness recovery percentage, it is quite complicated to be accomplished since it requires the removal of the internals and of the primary coolant in order to place the heating source. Moreover, dry annealing requires ensuring that other parts of the plant, e.g. the reactor cavity concrete, are not harmed by the high temperatures reached.

Concerning the duration of the annealing treatment, Mager et al. (referenced by [7]) concluded that an excellent recovery of toughness-related properties could be achieved by annealing at 450ºC for 168 hours. After the treatment, the reembrittlement caused by irradiation follows the same rate as that experienced before annealing. For this reason, the increase of DBTT with fluence is often referred as "lateral shift", since the effect of annealing is simply to laterally shift the DBTT vs fluence curve towards higher fluence levels, as illustrated in Figure 8.

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Regulatory Guide 1.162 states that: "*Although thermal annealing has not yet been applied to a U.S. commercial power reactor, it has been successfully applied to other reactors. Two reactor vessels that have been successfully annealed are the Army's SM-1A in 1967, and the BR-3 in Mol, Belgium, in 1984. Both of these reactors operated at temperatures low enough to permit "wet annealing" at a temperature of 650F using the reactor coolant pumps as the heat source. In addition, at least 12 Russian-designed VVER-440 PWRs, which operate at conditions similar to U.S. PWRs, have been annealed in Russia and Eastern Europe at temperatures of approximately 850F, using dry air and radiant heaters as the heat source*" ([7]).

5. Current regulations on RPV embrittlement

Since RPV irradiation can not be completely avoided, the occurrence of embrittlement in the beltline region is, at some extent, known to occur. Therefore, while the need to reduce the level of embrittlement is an issue addressed during the reactor design phase, most of the related regulations are aimed at preventing the operation of the reactor in conditions that, by virtue of the experienced embrittlement and of the possible presence of a crack, may lead to brittle fracture and therefore RPV failure. The requirements for brittle fracture prevention are discussed in several official documents, the most important of which are summarized as follows.

5.1 Appendix A of 10 CFR Part 50

Appendix A, "General Design Criteria for Nuclear Power Plants", of 10 CFR Part 50 ([4]), establishes general recommendation to prevent RPV failure, In fact, General Design Criterion 31 states: "*The reactor coolant pressure boundary shall be designed with sufficient margin to assure that when stressed under operating, maintenance, testing, and postulated accident conditions (1) the boundary behaves in a nonbrittle manner and (2) the probability of rapidly propagating fracture is minimized(...)*"([4]). Criterion 31 also requires RPV design and operation to account for material property modifications resulting from irradiation, e.g. embrittlement.

5.2 Appendix G of 10 CFR Part 50

The requirements contained in this Appendix are expressed in terms of:

- minimum allowed toughness of the RPV in the beltline region (102 J before irradiation and 68 J throughout the entire RPV life);

- pressure and temperature limits for the various operating phases that a reactor experiences during its design life, including unanticipated operating events.

In expressing limits to pressures and temperatures, Appendix G of 10 CFR Part 50 ([4]) refers, as a basis, to the requirements established by Appendix G of ASME Code [8]. To these requirements, Appendix G of 10 CFR Part 50 ([4]) applies additional conservativisms, which essentially consists of an increase of the minimum allowed RPV temperature by an amount ranging between 20 and 70ºC, depending on the operating phase under consideration (core critical or subcritical, high pressure or low pressure, fuel in the vessel or not).

Appendix G of 10 CFR Part 50 ([4]) does not discuss the methodology required to decide if a certain pressure-temperature condition allows the RPV to operate in safe-mode, i.e.

without hazard of brittle fracture. Instead, it explicitly refers to the ASME Code, whose most important points are discussed below.

5.2.1 ASME Code: pressure and temperature limits during normal operation

From the operating condition viewpoint, the effect of the embrittlement is to constrain the number of possible combinations pressure-temperature (*p-T*) that each point of the RPV can tolerate without experiencing nonductile propagation of a preexisting crack assumed to exist at that point.

It is well known that the brittle crack propagation is governed by the stress intensity factor at the crack edges. When such a factor exceeds a critical value K_{Ic}, referred as Plain Strain Fracture Toughness[4], the propagation occurs. K_{Ic} depends on the type of crack and in particular on the actual toughness of the material, i.e. the toughness calculated accounting for the irradiation effect. At any time during the RPV operating life, i.e. at any given level of fluence, the verification of whether a *p-T* combination can be tolerated at a certain location in the vessel is performed by comparing an ad-hoc defined stress intensity factor K_I^*, which is function of *p* and *T*, to the lower bound of experimental measures of fracture toughness performed on specimens made of the same material of the RPV and irradiated to the same level of fluence. The following inequality must be satisfied:

$$K_I^* < K_{Ic} \tag{4}$$

Figure 9 shows the curve used to perform this comparison. The parameter plotted is the experimental Plain Strain Fracture Toughness, which was measured at different temperatures for various specimens, each irradiated to a certain level of fluence, and therefore having a certain DBTT. It is important to note that, "*no available data points for static tests fall below the curve*" (Appendix G of [8]), i.e. it has never been observed a

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[4] Fracture toughness, K_c, is a quantitative way of expressing "*a material's resistance to brittle fracture when a crack is present*" ([9]). It represents the minimum stress intensity factor that, once originated in a specimen at the edges of a preexisting crack, causes the crack to propagate, yielding brittle fracture. This parameter depends on several factors, among which the dependences on the material toughness (via T-DBTT) and on strain rate $\dot{\varepsilon}$ are usually graphically (and not analytically) represented. For a given temperature T and $\dot{\varepsilon}$, K_c is defined as: $K_c = Y \cdot S \sqrt{\pi \cdot a}$, where S is the stress intensity, *a* is a function of the size and type (internal, edge, etc.) of the preexisting crack, while *Y* is a dimensionless factor depending on the type of crack and on the ratio of crack size to specimen width (measured in the direction of crack propagation). It is important to note that, for very large values of this ratio, i.e. for large specimen width *b*, *Y* looses its dependence on the cited ratio, tending to a constant value which depends only on the type of crack. Because of the relation above, such independence of *Y* from the specimen dimensions translates into an independence of K_c from the specimen dimensions. In fact, K_c decreases as *b* increases, but remains constant and equal to K_{Ic}, referred as Plain Strain Fracture Toughness, for widths larger than a critical width usually indicated as B. It has been experimentally verified that, as a general rule, $B \geq 2.5 \left(\dfrac{K_{Ic}}{\sigma_y} \right)^2$. Since for a typical ferritic steel ($\sigma_y = 350 MPa$), K_{Ic} varies from about 35 MPa m$^{0.5}$ (at T=DBTT-50ºC) to about 200 MPa m$^{0.5}$ (at T=DBTT+50ºC), the resulting values for B ranges between few cm to ~80 cm. Being the typical RPV wall thickness well below 80 cm, the RPV resistance to brittle crack propagation is actually larger than K_{Ic}. Therefore, the use of the Plain Strain Fracture Toughness K_{Ic} to characterize the RPV resistance to brittle fracture can be defined as a conservative estimate of its actual ability to withstand nonductile fracture.

brittle crack propagation occurring for stress intensity factors lower than those represented by the curve.

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As mentioned earlier, the acceptability of a given pressure-temperature combination requires the calculation of an ad-hoc defined stress intensity factor, K_I^*, which needs to be compared to K_{Ic}. Consistent with the definition of stress intensity factor, K_I^* depends on the actual level of stress, on the RPV thickness and on the characteristics of the crack. However, the crack characteristics are not chosen to represent a real crack that may be present in the RPV walls, but they are instead conservatively referred to the so-called "maximum postulated cracks". For most RPV locations, the postulated defects are axially-oriented surface defects[5], both on the inside and on the outside surface. Their size depends on the thickness t of the RPV section under investigation, as follows (Appendix G of [8]):

- t<102 mm: crack depth of 25 mm;
- 102<t<305 mm: crack depth of 0.25t; crack length of 1.5t;
- t>305 mm: crack depth of 76 mm; crack length of 457 mm.

Once the maximum postulated crack has been defined, K_I^* is calculated as[6]:

$$K_I^* = 2K_{Im} + K_{It} \tag{5}$$

where K_{Im} is the membrane tension stress intensity factor while K_{It} is the thermal stress intensity factor, both referred to the location of the RPV under investigation. The former is calculated as:

$$K_{Im} = M_m \frac{pR_i}{t} \tag{6}$$

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[5] Cracks are assumed to be circumferentially oriented in the study of circumferential welds.
[6] When locations near flanges and nozzles are investigated, a bending term is added to the right hand side of equation (5).

where t is the thickness of the RPV section under examination, p is the internal pressure, R_i is the vessel inner radius and M_m is a function of t and of whether the crack is on the inside or on the outside surface (Appendix G of [8]). The thermal stress intensity factor is instead the maximum stress intensity factor that generates in the RPV because of the through-wall thermal gradient produced during heatup and cooldown phases. Being K_I^* dependent on the heatup/cooldown rate, the coolant temperature variation with time can not take any value, but is typically limited at 56 ºC/hr.

By introducing (5) and (6) into (4), the maximum allowed pressure at a given time during the RPV operating life can be calculated as:

$$p = \min\left\{ \left[\frac{K_{Ic} - K_{It}}{2M_m} \left(\frac{t}{R_i} \right) \right]_j \right\} \tag{7}$$

where the subscript "j" indicates the j-th geometric location in the RPV.

It is important to note that, besides the conservativism implicit in the use of the "maximum postulated cracks", additional conservativisms characterize the procedure recommended for the verification of the allowable pressure. The use of a factor "2" in equation (5) together with the conservative choice previously described for the development of the K_{Ic}-curve are good examples of such an extra-conservativism. For this reason, while discussing the concept of "maximum postulated cracks" Appendix G of [8] states that: "*the prevention of nonductile fracture is ensured (…) even if the defects were to be about twice as large*".

5.2.2 ASME Code: p and T limits during unanticipated operating events

Appendix E of the ASME Code ([8]), provides acceptance criteria on the maximum allowed pressure during two types of unanticipated operating transients: pressurized thermal transients, in which the coolant temperature variation rate is larger than 5.6 ºC/hr, and isothermal pressure transients in which this rate is less than 5.6 ºC/hr. During the former, the pressure must not exceed the design pressure, while the coolant temperature must be at least 31 ºC above the RPV DBTT. Instead, during the more frequent isothermal pressure transients, the maximum pressure must not exceed the allowable values listed in Table 1 (from Table E-2 of Appendix E of [8]):

Table 1: Maximum allowable pressure as a function of T-DBTT (for design pressure grater than 2400 psig)		
Type of unanticipated operating event	T-DBTT, ºC	Maximum allowable pressure, psig (MPa)
Pressurized thermal transients	≥31	Design pressure
Isothermal pressure transients	13.9	1.1×design pressure
	8.3	2400 (16.548)
	5.5	2250 (15.513)
	0	2000 (13.790)
	-5.5	1750 (12.066)
	-13.9	1500 (10.342)
	-27.8	1200 (8.274)
	-41.6	1000 (6.895)
	-58.3	850 (5.860)
	-92.2	800 (5.516)
	-111.1	750 (5.171)

General note: linear interpolation is permitted

5.3 Appendix H of 10 CFR Part 50

This Appendix, which is based on the ASTM Code Section "Standard Recommended Practice for Surveillance Tests for Nuclear Reactor Vessels" ([10]), regulates surveillance programs aimed at monitoring changes in the fracture toughness properties of the RPV beltline region. Besides the points discussed in detail by the ASTM Code, this Appendix requires that:

- <u>no material surveillance program is required for those reactors whose RPV accumulates, at the end of its design life, a fast neutron fluence below 10^{17} n/cm^2: this value can be therefore considered the damage threshold for neutron-induced embrittlement;</u>

- surveillance specimen capsules must be located in the beltline region so that they can duplicate the neutron spectrum, the temperature history and the maximum neutron fluence experienced by the RPV;

- the integrated surveillance program, i.e. representative materials chosen for surveillance of a reactor are irradiated in other reactors, is allowed as long as there is a demonstrated high degree of similarity among the reactors involved in the integrated program.

5.4 Section 50.61 of 10 CFR Part 50

This document establishes requirements aimed at preventing the occurrence of the most well known embrittlement-induced accident event, i.e. the Pressurized Thermal Shock event (PTS). Because of its singular nature, and because of its catastrophic outcome, PTS is described separately in the next section, together with the related regulations.

6. Pressurized Thermal Shock Event

In unirradiated conditions, the ferritic steels of which LWR vessels are made have DBTT ranging between -30 and 0 ºC ([3]). In absence of embrittlement phenomena, this low temperature would prevent the RPV from experiencing brittle behavior since the operating temperatures are much higher. However, the DBTT resulting from the fast fluence absorbed over a 40-60 year period, which can be up to 6-8×10^{19} n/cm^2 for a typical PWR, can reach values up to 120 ºC (see Figures 5 and 6). This value is not far from temperatures that the RPV may experience during cooldown transients. If these temperature transients occur at low pressure, there is not significant risk of RPV failure, in that RPV walls are subjected to relatively low stresses. However, transients may occur during which the RPV is still at high pressure or it repressurizes while its walls experience a rapid reduction in temperature due to cold water injection. This type of transient, known as Pressurized Thermal Shock, may yield a state of stress high enough to cause, in presence of an initial crack, RPV failure. In case of cold water injection, which is expected for many severe accidents, the importance of reducing the internal pressure can be shown by simply saying that the maximum cooling rate typically allowed by the plant operating procedures, i.e. 56ºC/h, if experienced at 2250 psia yields a stress intensity factor 3-4 times higher than that characterizing a depressurization transient ([3]). Although one severe cooling event at pressure has already occurred (Rancho Seco PWR, see Figure 10), a recent study showed that "*US PWRs do not approach the levels of embrittlement to make them susceptible to PTS failure, even during extended operation*

well beyond the original 40-year design life". In fact, "*the simultaneous occurrence of crtitical-size flaws, embrittled vessel, and a severe PTS transient is a very low probability event*" (NUREG-1808, [11]).
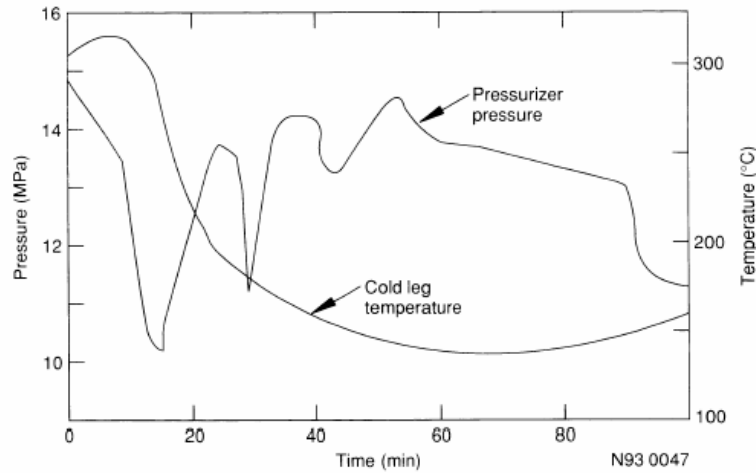

Figure 10: Pressure and temperature variations during the 1978 accident at Rancho Seco ([3])

6.1 PTS prevention regulations

Section 50.61 of 10 CFR Part 50 ([4]) is aimed at monitoring the level of RPV embrittlement in order to specifically prevent PTS occurrence. The monitoring process is extremely important bacause, only for PTS-related purposes, the neutron fluence of interest is that corresponding to the expiration date of the operating license. Based on the maximum allowed fluence for PTS prevention, the continuous monitoring of the level of fluence allows to "plan" the future neutron flux history on the RPV walls, so that a certain safety margin is ensured at the end of the operating license and, at the same time, the fuel loaded in the peripheral assemblies can be completely burnt. In other words, the fluence monitoring allows satisfying safety and economic requirements simultaneously.

The above mentioned "maximum allowed fluence for PTS prevention" is expressed, in Section 50.61 of [4], in the form of the DBTT that is expected to characterize the RPV beltline region at the end of the operating license, DBTT$_{PTS}$. The "PTS Screening Criterion" establishes that the maximum allowed DBTT$_{PTS}$ is 132ºC for plates, forgings and axial weld materials, and 149ºC for circumferential weld materials ([4]). The verification of whether these limits are exceeded or not is performed using the following relation for *DBTT$_{PTS}$*:

$$DBTT_{PTS} = DBTT_U + M + \Delta DBTT_{PTS} \tag{8}$$

where *DBTT$_U$* is the Ductile Brittle Transition Temperature of the unirradiated material, *M* is a positive factor accounting for uncertainties in the value of *DBTT$_U$*, in the material composition, fluence and on the calculation procedures. Finally, *ΔDBTT$_{PTS}$*, which can be calculated using equation (1), is the increase in Ductile Brittle Transition Temperature experienced by the material at the end of RPV operating license.

It is interesting to note that the value of DBTT$_{PTS}$ calculated using equation (8) "*will cause about five US plants to exceed the PTS screening criteria limits before the expiration of their 40-year operating licenses, and four more will be within 1ºF of the screening criteria limits*" ([3]). In these cases, in order to avoid premature retirement of

the RPV, Section 50.61 of [4] allows the RPV beltline to be given a thermal annealing treatment so that toughness and DBTT can be recovered and the PTS screening criterion be satisfied.

7. Limitations and flaws of current regulations

The level of conservativism characterizing the current regulations is satisfactory as long as a high fast neutron flux is assumed to play the major role in causing RPV toughness reduction and DBTT increase. Such an assumption is undoubtedly true for those reactors, especially Generation III PWRs and WWERs, for which a relatively high fast neutron flux at the RPV beltline is not only the cause of a significant embrittlement, but is also the preeminent cause of this effect. In other words, the current regulations well apply to those extreme cases which led, decades ago, to the awareness of the existence of a real hazard for the RPV integrity. However, not all the reactors, even within the Generation III family, are characterized by a high fast neutron flux at the RPV beltline, and this becomes even more true when the attention is shifted to advanced reactors, which were far away from the attention of the regulators at the time the regulations were issued.

The inadequacies of the current regulations are essentially two, which can be distinguished in a "flaw" and a "limitation". The former arose from the analysis of the material cut from the RPV of the German Gundremmingen BWR, while the latter from studies performed on the High Flux Isotope Reactor (HFIR) and applicable to the ABWR. They are discussed in the following Sections.

7.1 The "flaw": the underrated importance of flux effect

As discussed so far, and as very well known, the RPV embrittlement is always put in relation with the time-integrated fast neutron flux, i.e. the fast neutron fluence. As a consequence, the level of embrittlement predicted for the RPV beltline region at the expiration of the operating license depends exclusively on the total number of fast neutrons that have collided against the RPV wall during 40-60 years of operation. Thus, the time history of the irradiation does not play any role, and that a 10^{19} n/cm^2 fluence has been accumulated over 40 years or 40 days does not affect the result. Consistent with this assumption, several embrittlement-prediction methods are based on experimental data coming from test reactors where the fast neutron flux is orders of magnitude higher than that experienced by commercial LWR vessels.

The analysis of "trepan", the material cut from the RPV of the German Gundremmingen BWR decommissioned in 1977, revealed that embrittlement had proceeded faster than expected, even exceeding the embrittlement level of a test reactor characterized by fast neutron fluence 4 times higher ([12]). Figure 11 shows the results of Charpy impact tests for unirradiated material, for trepan, and for specimens from a test reactor. The significantly low Upper Shelf Energy for trepan is quite evident.

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The reason of this apparent inconsistency lie just in neglecting, when the fast neutron flux is relatively low like in BWRs, the flux history that has led to the observed final value of fast neutron fluence. A possible explanation was given by H. Ino, professor in the Department of Mechanical Engineering at Hosei University, Japan ([12]). By means of a computer simulation based on a chemical rate equation theory, he investigated the effect of different values of fast neutron flux on the increase of yield stress, which is one of the consequences of neutron irradiation (see Figure 3). He also performed a sensitivity analysis by changing the copper content, based on the well known embrittlement-enhancement effect due to this alloy element. It is crucial to note that the simulation was performed with a constant fluence, but varying the neutron flux. The results are summarized in Figure 12 where, for the purposes of this discussion, only the continuous lines need to be considered. The parameter on the x-axis is the damage rate expressed in displacement per atom per second (dpa/s), which can be interpreted as the fast neutron flux (not fluence!) in view of the approximate correspondence: 1 dpa ~ 7×10^{20} n/cm^2 ([12]). It can be noticed that the level of embrittlement, in the form of change of yield stress with fixed fluence, is higher at lower fluxes. In particular, for a low fast flux reactor like a BWR, the level of embrittlement that would be predicted by the current methodologies and regulations is that corresponding to harder fluxes, i.e. PWR-like fluxes and even harder (from test reactors), resulting in an evident underestimate of the embrittlement. Although this difference increases as the copper content increase, the underestimate corresponding to typical RPV Cu contents (below 0.3%) is not negligible.

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The importance of the neutron flux in determining the level of embrittlement has been also observed by comparing two types of specimens extracted from Japanese BWRs: those located very close to the core and therefore subjected to a neutron flux larger by an order of magnitude, and those located on the RPV walls. In Figure 13 they are referred as "BWR accelerated" and "BWR" respectively. Figure 13 also shows the calculated profile for very embrittlement-sensitive steels (worst case), embrittlement-sensitive steels, and embrittlement-insensitive steels. It can be noticed that, in spite of absorbing a lower fluence, the specimens located at the BWR vessel walls (full dots) show the same ΔDBTT as those located closer to the core and thus absorbing higher fluence (other than higher flux). This is again the low-flux embrittlement enhancement phenomenon investigated by Ino.

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7.2 The "limitation": the neglected gamma-ray effect

While speaking about reactions causing embrittlement and <u>not</u> involving fast neutrons, Mansur and Farrell ([1]) state: *"special circumstances may be encountered where these reactions become significant or even dominant with respect to fast neutron-induced"* reactions. As discussed in Section 2.2, in order that gamma-ray induced embrittlement can be neglected with respect to fast neutron induced embrittlement, γ-ray flux must be less than 100 times the fast neutron flux. In Generation III LWRs, this condition is satisfied, and therefore the methodologies used to predict the level of embrittlement, which are neutron-based, are applicable. However, for some advanced and GenIV reactors, this may not be true. In fact, the ratio between the gamma flux Φ_γ and the total (fast + thermal) neutron flux Φ_n reaching the RPV walls varies significantly depending on the width of the water gap separating the core from the RPV walls. This is due to the shielding effect caused by this gap, which is stronger for neutrons than for γ-rays since the latter are less effectively attenuated. As a consequence, the thicker is the water gap the larger is the ratio Φ_γ/Φ_n at the RPV wall. This point was arisen by General Electric for the Advanced Boiling Water Reactor (ABWR) ([13]), which is provided with a ~900 mm wide water gap in order to accommodate ten internal recirculation pumps. Such a wide gap makes the ABWR vessel less susceptible to embrittlement since the neutron flux incident on the RPV walls is about $1/10^{th}$ of that characterizing a BWR/6. Simultaneously, however, the mentioned unequal water shielding effect causes a significant increase of the fraction of damage caused by gamma irradiation, which reaches 60% of that caused by neutrons at a quarter of the wall thickness, and even 110% at the wall inner surface (where $\Phi_\gamma/\Phi_n \sim 3000$, see Figure 14a) ([13]). Figure 14b shows the dependence of the ratio of gamma-induced displacements to neutron-induced displacements on the water gap width, at a radial position corresponding to a quarter of the ABWR vessel thickness. In the same plot the points corresponding to a typical PWR and a typical BWR are also shown, so that the unusual situation characterizing the ABWR appears even more clearly.

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Oak Ridge National Laboratory ([1]) proved that, in presence of large water gaps, the use of data coming from neutron-dominated-embrittlement tests to predict RPV damage yields a significant overestimate of the time needed to reach a certain level of embrittlement. In fact, the term "accelerated embrittlement" was coined to indicate the early embrittlement of the coupons analyzed during the High Flux Isotope Reactor (HFIR) surveillance program ([14]). Like the ABWR, also the HFIR has a large water gap (~600 mm wide) interposed between the core and the RPV. The analysis of the surveillance data showed that HFIR specimens irradiated at fast neutron fluences of the order of 10^{17} n/cm^2 showed a DBTT increase of tens of degree C, when instead experimental measures performed in test reactors not provided with large water gaps indicated that no embrittlement should occur at fast fluences below about $6{\times}10^{17}$ n/cm^2. In the HFIR specimens a significant γ-ray effect was in fact superposed to that of fast neutrons. Figure 15 shows the HFIR data compared to those coming from typical test reactors.

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8. Summary and conclusions

The most important points arising from the discussion on embrittlement can be summarized as follows:

1) RPV embrittlement is a phenomenon of mechanical property degradation that in principle can be caused by thermal neutron, fast neutron and gamma-ray irradiation, other than nuclear reactions, usually neutron-induced, resulting in He production inside the RPV walls. The contribution percentages due to each of the mentioned causes strongly depend on the flux associated to each: in fission reactors, for the same flux, fast neutron contribution dominates over thermal neutron (by about two orders of magnitude) and over gamma-rays (by about three orders of magnitude). However, while the dominancy over thermal neutrons can be reasonably assumed in all reactors, circumstances may occur in which the gamma-ray flux at the RPV is significantly higher than fast neutron flux, resulting in an overturning of the mentioned contribution percentages.

2) For the same type and intensity of irradiation, the embrittlement level increases as the irradiation temperature decrease.

3) Alloy elements such as Cu and Ni have an enhancement effect on embrittlement.

4) For a given RPV steel at a given temperature, current regulations recommend to predict the severity of embrittlement by using a time-integrated fast flux model, in which the embrittlement indicator parameters are expressed as a function of the fast neutron fluence only. This model is conservative as long as gamma-ray contribution to embrittlement is negligible (γ-ray flux<100×fast neutron flux) <u>and</u> the fast neutron flux at the RPV wall, Φ_{nf}, is relatively high. The concept of "relatively high" has not been precisely defined yet, but it is reasonable to say that the fast fluence model can be applied to Gen III PWRs ($\Phi_{nf} \sim > 3 \times 10^8$ n/cm^2 s), while it is typically not conservative in BWR conditions ($\Phi_{nf} \sim 10^7$ n/cm^2 s).

5) When gamma-ray flux at the RPV walls is less than 100 times the fast neutron flux, RPV embrittlement can be reasonably considered a threshold phenomenon with respect to fast neutron fluence. The threshold below which embrittlement is negligible, for typical LWR RPV irradiation temperatures, is 10^{17} n/cm^2.

6) The recent findings concerning:

   - the unexpected high dependence of embrittlement on fast neutron flux at low fluxes, and

   - the water-gap effect in enhancing the gamma-ray contribution to embrittlement;

   coupled with the current tendency of reactor design in:

   - decreasing the fast neutron flux at the RPV wall, and

   - interposing, in some cases, large water gaps between core and RPV

   require a revision of the current fast-fluence-based regulations.

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