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# 12 Due to Neutron

### Degradation Due to Neutron Embrittlement of Nuclear Vessel Steels: A Critical Review about the Current Experimental and Analytical Techniques to Characterise the Material, with Particular Emphasis on Alternative Methodologies

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### 1. Introduction

The pressure vessel constitutes the most important structural component in a nuclear reactor from the point of view of its safety. The core of the reactor, that is, the nuclear fuel, is accommodated inside the vessel. This material is composed of fissile nuclides that undergo chain nuclear reactions of an exothermic nature, thus generating usable energy. Uranium 235 (U-235) is the only isotope in Nature which is fissile with thermal neutrons; hence, it is used as nuclear fuel in Light Water Reactors (LWRs). Two technologies of LWRs can be distinguished, Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs). Currently, more than 400 nuclear reactors operate in the world of which, approximately, 57% are PWR and 22% BWR. The original design lifetime for LWRs is 40 calendar years; nevertheless, the current target for most plants in many countries in Europe, Japan and the USA is to extend their operative lifetime up to 60 years.

The nuclear vessel is a virtually irreplaceable element which is subjected to operating conditions that lead to a progressive degradation in time of its constituent steel. The chain fission reactions of U-235 entail the emission of high energy neutrons that inevitably impact the internal surface of the vessel. These collisions give rise to a complex series of events in the nano and microstructural scale that, in the end, modify the mechanical properties of the steel leading to its embrittlement, that is, the decrease in its fracture toughness. This phenomenon is most intense in the so called beltline region (which is the general area of the reactor vessel near the core midplane where radiation dose rates are relatively high). The total number of neutrons per unit area that impact the internal surface of the vessel represents the neutron fluence; in practice, only that fraction of the energy spectrum corresponding to a neutron kinetic energy higher than 1 MeV is considered to be capable of triggering damage mechanisms in the vessel steel; these neutrons are referred to as fast neutrons. Typical design end of life (EOL) neutron

fluences (E>1 MeV) for BWRs are in the order of  $10^{18}$  n/cm<sup>2</sup>, whereas for PWRs this number is about  $10^{19}$  n/cm<sup>2</sup>.

In Section 2 of this chapter, the embrittlement of nuclear vessel steels is described from a purely phenomenological perspective as well as from the point of view of the legislation currently in force. The phenomenon of the ductile to brittle transition and the influence of embrittlement on it are particularly stressed. In Section 3, a brief description of the main characteristics of the nuclear power plants surveillance programmes is presented; the requirements that they envisage as well as the information that they allow to be obtained are pointed out. The physical mechanisms that take place in the nano and micro levels leading to the material embrittlement are detailed in Section 4 where a brief exposition concerning the most relevant predictive models for embrittlement is also presented. Finally, in Section 5, the promising method of the Master Curve is described; this represents an improved methodology for the description of the fracture toughness of vessel steels in the ductile to brittle transition region, susceptible to be incorporated in the current structure of the surveillance programmes.

# 2. The embrittlement of nuclear vessel steels and its influence on the ductile to brittle transition region: Phenomenology and regulations

# 2.1 The phenomenology of the embrittlement and the ductile to brittle transition region

As mentioned above, neutron irradiation reduces vessel steel toughness. To understand the concept of material fracture toughness, Fracture Mechanics theory must be referred to. In this section, the basic principles of Fracture Mechanics are briefly presented. Material toughness can be understood as its resistance to be broken when subjected to mechanical loading (forces, stresses) in the presence of cracks / flaws (that is, a sharpened discontinuity). Traditionally, RPV toughness variation has been indirectly quantified by means of Charpy impact tests. These tests were introduced by the French researcher George Charpy in 1905. The preparation, execution and interpretation of Charpy tests are currently regulated by the ASTM E 23 standard (ASTM E 23, 2001). The test consists of breaking a small dimension (10 x 10 x 55 mm<sup>3</sup>) specimen by the impact of a pendulum released from a controlled height and measuring the height it achieves after the breaking (see Figure 1). First, a notch has to be machined in the specimen that acts as stress concentrator, forcing the fracture process. The expression "notch" refers to a blunted defect and, therefore, must not be misunderstood as a crack or flaw (described above). The main numerical result provided by the Charpy test (though not the only one) is the energy absorbed during the specimen breaking, also called resilience, which is used as a semi-quantitative estimation of the material fracture toughness.

Because of its simplicity of execution and for historical reasons, this experimental technique is still employed in several industries such as the naval, building and pressure vessel design and, in particular, in nuclear vessels. However, several limitations associated to the Charpy impact test can be mentioned. First, the fracture process occurs under highly dynamic conditions and, therefore, can hardly be extrapolated to quasi-static fracture processes (as the ones occurring in many components and, predictably, in nuclear vessels). Moreover, it has been demonstrated that the results provided by the Charpy test, such as the resilience, do not represent genuine material properties, which can be applied to components of different dimensions and loading conditions; as mentioned above, the resilience may be Degradation Due to Neutron Embrittlement of Nuclear Vessel Steels: A Critical Review about the Current Experimental and Analytical Techniques...

considered only as a semi-quantitative estimator. Finally, the standard specimen notched geometry, which facilitates its breaking, is far from simulating an ideal crack, that is, a sharpened discontinuity embedded in the material.

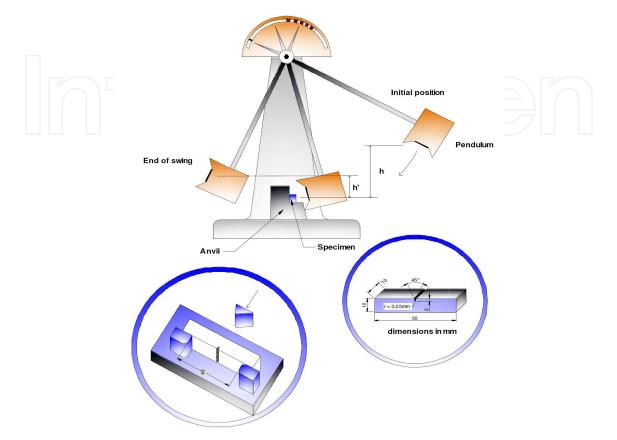


Fig. 1. Sketch of the Charpy pendulum and the standard Charpy specimen

Experience shows that the vessel steel fracture properties drastically depend on temperature. This phenomenon is clearly appreciated in the resilience curves, which are obtained from Charpy tests carried out at different temperatures. Thus, in Figure 2, it can be appreciated how the absorbed energy in the specimen breaking process varies from low values at low temperatures, in the region called brittle or Lower Shelf, to high absorbed energies at high temperatures, in the ductile region or Upper Shelf. The intermediate temperatures constitute what is called the Ductile to Brittle Transition Region, DBTR. One additional property of the DBTR is that the resilience values present an important scatter, as can also be inferred in Figure 2. In order to simplify the analysis, it is common practice to fit the data according to mathematical expressions such as the hyperbolic tangent, see Figure 2 (the free parameters A, B, C and D must be obtained through the fitting procedure, typically a least squares method). The existence of three regions and the scatter of the results in the DBTR are also observable in the fracture toughness curves. According to this, the description of the vessel steels toughness in the DBTR represents a remarkable engineering problem because, on the one hand, this property changes ostensibly with temperature and, on the other hand, it shows an important scatter.

Figure 3 shows an example of a vessel steel neutron irradiation embrittlement. In this figure, Charpy impact test results (absorbed energy vs. temperature) from unirradiated and irradiated material, respectively, are represented. As can be seen, the most remarkable effects are a shift

200 180 **UPPER SHELF** 160 Π 140 **TRANSITION** 120 3 REGION Energy ( 100 80 60 40 20 I OWER SHELE 0 -150 -100 -50 0 50 100 150 200 Temperature (°C)

of the curve in the DBTR towards higher temperatures accompanied by a reduction in the absorbed energy in the Upper Shelf, that is, a generalised embrittlement process.

Fig. 2. Influence of temperature on the results (absorbed energy) of the Charpy test; the data were fitted to a hyperbolic tangent curve

In regard to nuclear vessel structural integrity assessment, it is of great interest to determine the neutron irradiation effect on the fracture properties in the DBTR. In the nuclear industry, it is common to simplify this phenomenon by means of the definition of standard temperatures that are considered as representative. Among these, the most used ones are  $T_{28J}$  and  $T_{41J}$ , that is, the temperatures where the Charpy impact test absorbed energies are 28 J and 41 J, respectively (obtained after fitting the results to a hyperbolic tangent curve).

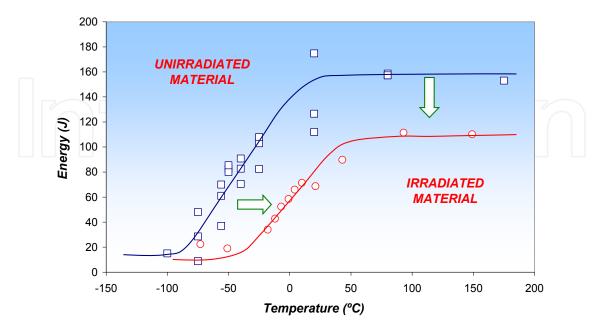


Fig. 3. Description of the influence of neutron irradiation on the Charpy curves (absorbed energy vs. temperature)

#### 2.2 The ASME curves and the reference temperature RT<sub>NDT</sub>

Since 1905, the year when George Charpy proposed his famous test, until the present, the knowledge of the mechanics that control the fracture process has experienced an important qualitative progress (a historic review can be found in the reference (Anderson, 1995)). Briefly summarising, at the end of the 50's the formulation of the so-called Linear Elastic Fracture Mechanics<sup>1</sup> (LEFM) discipline was available. LEFM accurately describes the fracture processes of brittle materials, that is, the materials that show a linear elastic response until failure (without the occurrence of previous relevant yielding). LEFM demonstrates that, the presence of an ideal crack in this type of materials when subjected to loading implies a stress state tending to infinite in the proximity of the crack front. On the other hand, it can also be demonstrated that the stress and strain fields, see Figure 4, in the proximity of the crack front depend exclusively on one parameter known as stress intensity factor (SIF or K<sub>I</sub>), which is a function of the applied load ( $\sigma$ ) and the defect size (a). It is worth mentioning that the subscript 'I' refers to the fracture mode I (tensile mode) as represented in Figure 4, which is the prevailing one in most cases of interest.

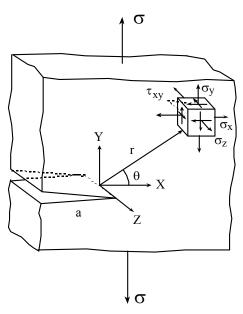


Fig. 4. Sketch of a cracked component / specimen and the stress state in the proximity of the crack front

Then, as  $K_I$  controls the stress-strain field in the fracture process zone (mode I), breaking will occur when  $K_I$  reaches a determined critical value, which will be dependent on the material. This reasoning provides a valid definition of the fracture toughness as the SIF critical value,  $K_{Ic}$ , and also the following fracture criterion (1):

$$K_{I}(\sigma, a) = K_{Ic}(material) \tag{1}$$

The SIF (and consequently the toughness  $K_{Ic}$ ) are usually expressed in MPa m<sup>1/2</sup>. Toughness  $K_{Ic}$  is determined following the rules of the ASTM E 399 standard (ASTM E 399, 2009) or some other equivalent procedure.

<sup>&</sup>lt;sup>1</sup> Thanks to the works of A.A. Griffith (Griffith, 1920), C.E. Inglis (Inglis, 1913), G.R. Irwin (Irwin, 1956, 1957) or H.M. Westergaard (Westergaard, 1939).

Unfortunately, LEFM is not valid for tough materials (such as reactor vessel steels), because these develop important yielding regions in the crack front before the occurrence of the breaking. In the 60's, several theoretical solutions describing the fracture processes in tough materials were proposed. The works of A.A. Wells (Wells, 1961) and J.R. Rice (Rice, 1968) allows the validity of LEFM to be extended, thus establishing the foundations of the socalled Elastic-Plastic Fracture Mechanics, EPFM. A new parameter, the J integral (which, like the SIF, depends on the loading state and the defect size) characterises the stress and strain fields in the crack front and, therefore, enables the fracture toughness to be defined generally, as the J integral limit value,  $J_c$  (sometimes also named  $J_{Ic}$  when it refers to the tensile fracture mode I). Also the following fracture criterion<sup>2</sup> can be presented (2):

$$J(\sigma, a) = J_c(material)$$
<sup>(2)</sup>

The J integral (and therefore the toughness  $J_c$ ) are usually expressed either in kJ  $\cdot$  m<sup>-2</sup> or in kPa  $\cdot$  m.

Then, since the 70's, once the Wells and Rice postulations were recognised, a more founded theoretical basis was made available to address material fracture, either brittle (LEFM) or ductile (EPFM). However, in the 60's, the period when Generation III reactors (LWR those covered in this text among them) started operating, EPFM was not available to the power plant design engineers. Among the different possible choices to address this problem, the one adopted by the USA authorities (represented by the Nuclear Regulatory Commission, NRC) is especially noteworthy. This regulation is contained in the ASME (American Society of Mechanical Engineers) Code.

The ASME International Boiler and Pressure Vessel Code (ASME Code) establishes rules of safety governing the design, fabrication and inspection of boilers and pressure vessels, and nuclear power plant components during construction. This standard is currently in force for USA designed pressure vessels. The ASME Code is made up of 11 sections and contains over 15 divisions and subsections. The Sections II (Materials), III (Rules for Construction of Nuclear Facility Components) and XI (Rules for In-service Inspection of Nuclear Power Plant Components) are of relevance for the contents of this chapter.

The researchers who elaborated the ASME code were aware of the intrinsic limitations of the LEFM; in particular, they knew that, a tough material breaks in a brittle manner (that is, in the LEFM range), only when large specimens are used. Thus, for example, the ASTM E 399 standard (ASTM E 399, 2009) includes requirement (3) on the thickness B of a toughness specimen in order to obtain a valid result according to the LEFM requirements.

$$B \ge 2.5 \left(\frac{K_{Ic}}{\sigma_{\gamma}}\right)^2 \tag{3}$$

where  $\sigma_{\rm Y}$  represents the material yield stress.

This possibility, however, is not applicable as the best solution to carry out the follow-up of the RPV steel because the space available inside the vessel is limited. Hence, it was decided to develop a different methodology, making use of Charpy impact specimens in the surveillance programmes (see Section 3) to subsequently correlate the results with the

<sup>&</sup>lt;sup>2</sup> Rigorously, this criterion describes the ductile fracture, that is, a fracture process with no stable propagation and non negligible yielding.

toughness values as defined by the LEFM principles. No doubt, this is an indirect and tortuous path but also the only one available to the ASME society researchers at that time. This decision, motivated by the theoretical limitations at the time it was made, is the cause of the conservatism of the current regulation.

Therefore, from the end of the 60's and the beginning of the 70's, an important experimental campaign was carried out, testing different vessel steels in the DBTR according to the LEFM stipulations. As explained above, in order to obtain representative results for tough steels (as those of nuclear vessels), the LEFM requires the employment of large specimens, which are difficult to be fabricated, handled and tested. The weight of some of these specimens reached a value of several tons. Due to the high cost of manufacturing, handling and testing these specimens, the reference curves were named as the "one million dollars curves". After compiling all the available data, it was appreciated that the curves obtained for the different steels showed, on the whole, a similar appearance; independently of the steel heat or composition, the most remarkable difference was its location on the temperature axis, see Figure 5. In consequence, the universal validity of the curves was established and a new parameter was defined for any sort of vessel steel, called reference temperature RT<sub>NDT</sub>, which allows its toughness-temperature curve to be located in the abscissa axis. Thus, representing in the abscissa axis the corrected scale (T – RT<sub>NDT</sub>), see Figure 5, all the different curves overlap in one single curve which is assumed to be the universal curve for the material behaviour.

In this sense, the toughness behaviour of the vessel material in the DBTR before irradiation (unirradiated material) is described by the reference temperature  $RT_{NDT(U)}$  which is obtained from a combination of the results of Charpy (ASTM E 23, 2001) impact and Pellini drop weight tests (ASTM E 208, 1995). Thus, this procedure implies correlating the fracture material behaviour from dynamic tests with uncracked specimens (only notched). This is not rigorously justified, either theoretically, or experimentally.  $RT_{NDT}$  is used to index two generic curves, developed in 1973, provided by the ASME Code relating toughness vs. temperature: the  $K_{Ic}$  curve describes the lower envelope to a large set of  $K_{Ic}$  data while the  $K_{IR}$  is a lower envelope to a combined set of  $K_{Ic}$ ,  $K_{Id}$  (dynamic test) and  $K_{Ia}$  (crack arrest test) data, being therefore more conservative than the former. Three important features can be appreciated: first, it is assumed that the ASME curves are representative of any vessel steel; second, in both curves, LEFM is considered; finally, the large scatter in the DBT region is removed by taking into account lower envelopes. Consequently, the method provides high conservatism in most cases.

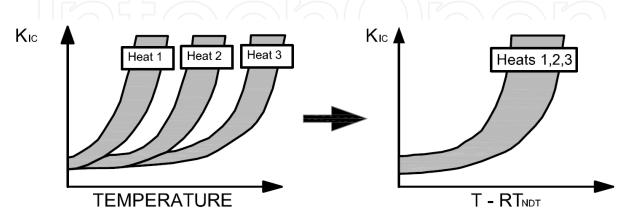


Fig. 5. Description of the process to obtain the ASME curves; definition of the reference temperature  $RT_{NDT}$ .

Figure 6 represents the ASME curves along with the experimental population which allowed their definition. The following equations (4, 5) are the mathematical expressions of the two curves:

$$K_{IC}(MPa \cdot m^{1/2}) = 36.45 + 22.766 \cdot e^{0.036 \cdot \left\lfloor T(^{\circ}C) - RT_{NDT}(^{\circ}C) \right\rfloor}$$
(4)

$$K_{IR}(MPa \cdot m^{1/2}) = 29.40 + 13.776 \cdot e^{0.0261 \cdot \left[T(^{\circ}C) - RT_{NDT}(^{\circ}C)\right]}$$
(5)

This procedure must be considered as a compromise solution, that is, an engineering tool with relevant limitations and inconsistencies. As will be mentioned in Section 5 of this Chapter, currently an alternative methodology, the Master Curve method, is available. This approach, besides offering a higher simplicity from the methodological and experimental point of view, also provides a complete robustness from the theoretical perspective. The Master Curve procedure enables a realistic follow-up of the surveillance programme vessel steel toughness evolution (see Section 3), avoiding the use of questionable correlations which unnecessarily penalise the representativeness of the subsequent structural calculations.

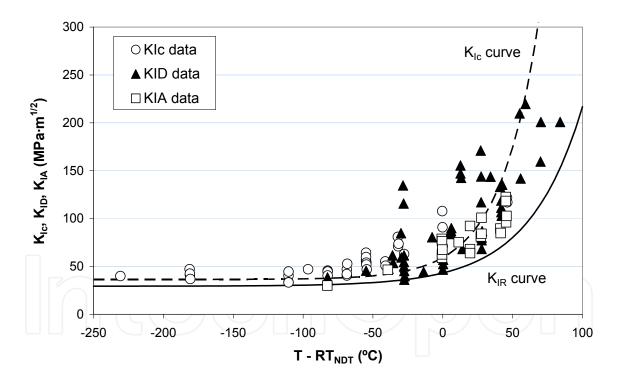


Fig. 6. ASME curves with the experimental data set used for their definition

#### 2.3 Influence of the neutron irradiation on the reference temperature $RT_{NDT}$

Although the physical mechanisms which lead to the vessel steel embrittlement will be extensively described in Section 4, it is considered convenient to include here some points in order to facilitate the comprehension of Section 3. The reduction of the material toughness due to neutron irradiation in the DBTR is currently estimated through semi-empirical methods based on the shift experienced by the Charpy impact curves obtained from the

surveillance capsule specimens (see Section 3). As stated in 10CFR50 (10CFR50, 1986), the effect of neutron fluence on the behaviour of the material is predicted by the Regulatory Guide 1.99 Rev. 2 (RG 1.99 (2), 1988) which provides equation (6) for the calculation of the evolution of  $RT_{NDT}$ :

$$RT_{NDT} = RT_{NDT(U)} + \Delta RT_{NDT} + M$$
(6)

where  $\Delta RT_{NDT}$  represents the shift in the reference temperature due to irradiation which is assumed to be equal to the shift of the Charpy transition curve indexed at 41J; thus,  $\Delta RT_{NDT} = \Delta T_{41J}$ . The third term, M, is the margin that is to be added to obtain a conservative estimation. The procedure in (RG1.99 (2), 1988) allows  $\Delta RT_{NDT}$  to be obtained even when no credible surveillance data are available by means of an equation based on the chemistry of the steel and the neutron fluence received (this issue is extensively described in Section 4).

#### 3. Surveillance programmes of nuclear power plants

For decades, the nuclear industry has known of the above described problems and, in consequence, has acquired the appropriate tools to be able to evaluate in advance the degradation of the vessel steel properties. Surveillance programmes are the tools currently employed to perform the follow-up of the evolution of the RPV steel properties throughout the operating lifetime of the reactor. These programmes consist of placing inside the reactor, from the beginning of plant operation, surveillance capsules containing specimens; these are made of a steel (base material, weld metal or heat affected zone material – see comments below regarding to this point) identical to the one which constitutes the vessel – along with flux wires necessary to estimate the neutron fluence and temperature gauges.

Surveillance capsules must be located within the reactor vessel, in the beltline, so that the specimen irradiation history duplicates as closely as possible, within the physical constraints of the system, the neutron spectrum, temperature history and maximum neutron fluence experienced by the reactor vessel. The fabrication history (austenitizing, quenching and tempering, and post-weld heat treatment) of the test materials will be fully representative of the fabrication history of the materials in the beltline of the reactor vessel. Because the capsules are located closer to the core than the inner vessel wall, the specimens suffer accelerated neutron fluence doses and, therefore, provide advanced information about the evolution of the embrittlement process. This phenomenon is described through the surveillance capsule lead factor (i.e., the ratio of the neutron fluence rate at the specimens in a surveillance capsule to the neutron fluence rate at the inner vessel wall, E > 1 MeV). It is recommended that the surveillance capsule lead factors has been selected to minimise the calculation uncertainties in extrapolating the surveillance measurements from the specimens to the reactor vessel wall.

Periodically, taking advantage of the plant refuelling outages, specimen removal is carried out (along with flux wires and temperature gauges) from inside the capsules. These specimens are subsequently tested, thus providing realistic information about the evolution of the material properties throughout the plant operating period. This is a method for assessing the material degradation caused by neutron irradiation. The issues related to the surveillance programme design along with the rules for the interpretation of the results of such programmes are

regulated in several ASTM standards (ASTM E 185, 2002; ASTM E 2215, 2002; ASTM E 853, 2001). Next, the minimum requirements considered in these standards for the design of a surveillance programme for monitoring the radiation-induced changes in the mechanical properties of ferritic materials in the beltline region are summarised; it is worth noting, for the sake of clarity, that the following list of characteristics is mainly focused on those aspects related with the material's behaviour; other issues, as the measurement of neutron fluence through dosimetry or the measurement of temperatures, are not considered of relevance for the purposes of the present contribution.

- It is recommended to obtain the baseline (i.e., unirradiated) data during the design of the surveillance programme. Alternatively, the specimens may be placed in secured storage for testing at the time the first surveillance capsule is withdrawn for evaluation.
- A minimum test programme shall include material taken from the following locations: (1) base metal from each plate or forging used in the beltline, and (2) each weld metal made with the same heat of weld wire and lot of flux and by the same welding procedure as that used for each of the beltline welds. The base metal used to fabricate the weld shall be one of the base metals included in the surveillance programme.
- Charpy V-notch specimens (corresponding to the Type A specimen described in Test Method E 23 ASTM E 23, 2001)) shall be used. Tension specimens of the type, size, and shape described in Test Method E 8 (ASTM E 8, 2009) are recommended. Fracture toughness test specimens shall be consistent with the guidelines provided in Test Methods E 1820 (ASTM E 1820, 2001) and E 1921 (ASTM E 1921, 2009).
- A minimum of 15 Charpy specimens shall be tested to establish a full transition temperature and upper- shelf curve for each material. It is recommended that at least six tension test specimens be provided to establish the unirradiated tensile properties for both the base metal and the weld metal. A minimum of two specimens at room temperature and two specimens at reactor vessel beltline operating temperature should be tested. The remainder of the specimens may be tested at intermediate temperatures as needed to define the effects of temperature on the tensile properties. It is recommended as well that a minimum of 8 fracture toughness specimens be tested to establish the reference temperature, T<sub>0</sub>, through Test Method E 1921 (ASTM E 1921, 2009) (see Section 5 for details about this issue) for the limiting beltline material and/or other fracture toughness tests be performed following Test Method E 1820 (ASTM E 1820, 2001).
- The first step in the implementation of a surveillance programme is the estimation of the peak EOL vessel inside surface fluence (EOL ID) and the peak EOL vessel 1/4-T (T being the thickness of the vessel) fluence<sup>3</sup> (identified as EOL 1/4-T). This data allows the transition temperature shift, ΔRT<sub>NDT</sub>, to be estimated. The reference temperature shift can be determined from the relationship found in Guide E 900 (ASTM E 900, 2002) (for details and alternative procedures, see Section 4 in this chapter).

<sup>&</sup>lt;sup>3</sup> The neutron fluence at any depth in the vessel wall attenuates as a consequence of the s cattering processes that occur between the neutrons and the crystal structure of the steel; these scattering processes are, in turn, responsible of the damage mechanisms leading to material's embrittlement. There are different models to predict this phenomenon, see, for example (RG 1.99 (2), 1988; ASTM E 900, 2002).

- A sufficient number of surveillance capsules shall be provided to monitor the effects of neutron irradiation on the reactor vessel throughout its operating lifetime. The basis for the number of capsules to be installed at the beginning of life is the predicted transition temperature shift,  $\Delta RT_{NDT}$ .
- The minimum number of tests on irradiated specimens (base and weld materials) for each irradiation exposure set (capsule) shall be as follows: 15 Charpy specimens, 3 tension specimens and 8 fracture specimens (only fracture toughness specimens from the limiting material are required. It is suggested that a greater quantity of specimens be included in the irradiation capsules whenever possible).
- The capsule withdrawal schedule should permit periodic monitoring of long-time irradiation effects. The suggested withdrawal schedule is provided in Table 1. At a minimum, the surveillance programme should consist of two priority 1 capsules (see Table 1) including samples of all irradiated materials (if there is no weld in the beltline, then there is no requirement to include weld material in the surveillance programme. Moreover, experience has shown that it is no longer necessary to include the heataffected zone material in the surveillance programme). The specimens in these capsules must be tested with target fluences corresponding to EOL-ID and EOL 1/4-T, respectively. If the estimated EOL  $\Delta RT_{NDT}$  is less than 56°C (100°F), the vessel embrittlement may be adequately monitored by testing only these two priority 1 capsules. The priority 2 capsule is required for plants that exceed this limit; this capsule should be scheduled for withdrawal early in the vessel life to verify the initial predictions of the surveillance material response to the actual radiation environment. Past practice for PWRs has shown that a target fluence of  $5.3 \ 10^{18} \text{ n/cm}^2$  (E>1 MeV) is adequate; a lower target fluence is appropriate for BWRs. The priority 3 capsule is included to better define the embrittlement trend in materials with large projected EOL ID  $\Delta RT_{NDT}$  values greater than 111°C (200°F).

Sequence	Target fluence	Priority
First	PWRs: 5 ·10 <sup>18</sup> n/cm <sup>2</sup> (E>1 MeV)	2 (required if $\Delta RT_{NDT} > 56^{\circ}C$ )
Second	EOL ¼-T	1 (required for all materials)
Third	EOL ID	1 (required for all materials)
Fourth	(EOL $\frac{1}{4}$ -T – 1 <sup>st</sup> Capsule)/2	3 (required if $\Delta RT_{NDT} > 111^{\circ}C$ )
Subsequent	Supplemental Evaluations	Not required

Table 1. Suggested Withdrawal Schedule

• Additional capsules may be needed to monitor the effect of a major core change or annealing of the vessel, or to provide supplementary toughness data for evaluating a flaw in the beltline or for monitoring the vessel over the extended lifetime. Supplementary testing may also be based on reconstitution of previously tested specimens following Guide E 1253 (ASTM E 1253, 1999).

### 4. The physical mechanisms of embrittlement, predictive models

The recognition of the importance of embrittlement processes led to the establishment of surveillance programmes as a strategy for monitoring the material properties of the vessel.

The limited data available from each specific monitoring programme is insufficient to predict the shift in the transition temperature. To get round this problem, the most widely used strategy is to turn to predictive formulas based on the statistical analysis of large populations of data from the monitoring programmes of several plants (Eason et al., 1998). The physical models developed in recent times include temperature, T<sub>i</sub>, fluence and the neutron flux with energies above 1 MeV,  $\Phi$  and  $\varphi$ , respectively, the contents of copper, nickel and phosphorus, and the form presented by the steel (plate, forging or welding). Recent studies have revealed minor influences associated with the presence of manganese and the heat treatment applied (Odette & Lucas, 1998). The phenomenon of the recovery of properties after an annealing treatment along with the subsequent embrittlement are scarcely documented at present and require further study. Given the large number of variables involved in the process, it is not feasible to elaborate predictive expressions of a purely empirical nature, particularly when these are to be extrapolated to high neutron doses. Fortunately, knowledge of the physical processes involved in the embrittlement enables reliable expressions to be proposed for estimating the shift in the transition temperature in terms of these variables.

It should be noted that all of the predictive equations for embrittlement used in the nuclear sector and gathered in the codes and regulations of each country are based on the results of the Charpy impact test. Most of these expressions were obtained for manganesemolybdenum-nickel steels, (typically used in the United States, Western Europe, Japan and chromium-molybdenum-vanadium Korea) or and chromium-molybdenum-nickelvanadium steels (used in the WWER reactors of Eastern Europe). These models express the embrittlement as a function of the presence of chemical elements (such as copper, phosphorus and nickel) and the value of fast neutron fluence. Sometimes, the irradiation temperature and the nature of the steel (weld or base material) are taken into account. It is also worth mentioning that, for most of the regulations, the fast neutron fluence is defined as the portion of the neutron spectrum with E>1 MeV, while in the Russian WWER reactors, the fraction with E > 0.5 MeV is used.

#### 4.1 Physical mechanisms of embrittlement

The mechanism of embrittlement is primarily based on the hardening produced by nanometric features that develop as a consequence of the impact of the neutrons on the crystal structure. The main embrittlement processes include (Odette & Lucas, 2001):

- Generation of lattice defects in displacement cascades by high-energy recoil atoms from neutron scattering and reactions. These primary defects are in the form of single and small clusters of vacancies and self-interstitials (Frenkel defects).
- Diffusion of primary defects also leading to enhanced solute diffusion and formation of nanoscale defect-solute cluster complexes, solute clusters and distinct phases, mainly copper-rich precipitates (CRPs).
- The presence of these nanofeatures can arrest the advance of the dislocations.
- As a consequence, this hardening process leads to the shift of the transition temperature detected experimentally, thus facilitating the material fracture through cleavages.

Obtaining a predictive expression to determine the transition temperature shift can be achieved by developing physical submodels of the above mentioned processes which depend on the metallurgical (Cu, Ni, P,...) and irradiation ( $\Phi$ ,  $\varphi$ , T<sub>i</sub>) variables (Eason et al., 1998; Odette & Lucas, 1998). The current understanding of the mechanisms of damage

production is essentially based on molecular dynamics (Stoller et al., 1997) and Monte Carlo numerical simulations (Wirth & Odette, 1999), combined with indirect experimental measurements. Thermodynamic-kinetic models are used to track the transport and fate of irradiation defects and solutes and to predict the number, size distribution and composition of the evolving nanofeatures. The characterization methods include: X-ray and neutron scattering, various types of electron microscopy, three-dimensional atom probe-field ion microscopy and positron annihilation spectroscopy. These researches allowed at establishing the following sequence (Odette & Lucas, 2001):

Neutrons create vacancies and interstitials, separated by some distance, by displacing atoms from their normal crystal lattice sites. The displacements are produced in cascades resulting from highly energetic primary recoiling atoms (PRA) generated by neutron scattering and reactions. The interaction of a high-energy neutron with an atomic nucleus results in significant energy transfer (R). For example, a 1 MeV neutron transfers up to about 70 keV to an iron PRA (Odette & Lucas, 2001). Part of the recoil energy is lost in the electronic cloud; hence, a lower kinetic energy is available for atomic collisions,  $R_d < R$ . The PRA kinetic energy is transferred by secondary, tertiary and n-subsequent generations of collision displacements, producing  $2^n$  recoiling atoms at lower energies ( $\sim R_d/2^n$ ). The process terminates when the kinetic energy of the nth-generation of recoils falls below that needed to cause additional displacements. On average, a PRA creates  $R_d/2D$  displacements, where  $D\sim 0.05$  keV. Thus, a typical value  $R_d = 20$  keV is able to produce a cascade of  $\sim 200$  displacements. Closely spaced interstitials and vacancies quickly recombine and only about one third of the initial displacements survive. Typically, this leaves a vacancy-rich cascade core, surrounded by a shell of interstitials.

Most of interstitials quickly cluster to form small, disc-shaped features that are identical to small dislocation loops. Along with the interstitials, these loops are very mobile. Diffusion of interstitials and loops within the cascade region causes additional recombination prior to their rapid long-range migration (unless they are strongly trapped by other defects or solutes). Although they are less mobile than the interstitials, vacancies also eventually diffuse. Through a series of local jumps, the vacancies and solutes in the cascade quickly begin to evolve to lower energy configurations, forming small, three-dimensional clusters, while others leave the cascade region. The small clusters are unstable and can dissolve by vacancy emission. However, the small clusters also rapidly diffuse and coalesce with each other, forming larger nanovoids, which persist for much longer times. Solute atoms bind to the vacancies and segregate to clusters. The vacancy emission rate is lower from vacancy-solute cluster complexes. Small solute clusters remain after all the vacancy clusters have been finally dissolved.

The neutron dose or damage exposure can be expressed in terms of displacements-per-atom (dpa) which partially accounts for the effect of the neutron energy spectrum. For a typical RPV neutron spectrum, an end of life  $\Phi = 3 \cdot 10^{19}$  n cm<sup>-2</sup> (E>1 MeV) produces about 0.1 dpa. However, most of the vacancies and interstitials eventually migrate and annihilate at sinks long distances from the cascade region. Thus long-range diffusion results in additional nanostructural evolution.

As a consequence of the above mentioned mechanisms three broad categories of nanofeatures can be distinguished:

• Copper rich or manganese-nickel rich precipitates (CRPs/MNPs).

- Unstable matrix defects (UMD) that form in cascades even in steels with low or no copper, but that anneal rapidly compared to typical low  $\Phi$  irradiation times.
- Stable matrix features (SMF) that persist or grow under irradiation even in steels with low or no copper.
- Grain Boundary Segregations (GBSs) of embrittling elements as phosphorus (Server et al., 2001).

Whereas the contributions of SMDs and CRPs imply the creation of obstacles blocking the displacement of the dislocations, the GBSs can reduce the intergranular cohesion thus modifying the mechanisms and properties of fracture. As a consequence, a change from transgranular to intergranular fracture can ocurr. This phenomenon has not been observed in USA vessels but in Brtitish (Server et al., 2001).

#### 4.2 Standard models for predicting the material embrittlement

This section includes a brief revision of the models proposed by the legislation currently in force that allows the embrittlement of the steel of the vessel to be determined throughout its operative lifetime. The vessels designed or manufactured in the USA are subjected to the regulations of that country. Specifically, the code of federal regulations 10CFR50 (10CFR50, 1986) is the reference document. According to this law, the Regulatory Guide 1.99 Rev. 2 (RG1.99 (2), 1988) establishes the procedure to determine the material embrittlement (see equation (6)); on the other hand, it points out that the plant surveillance programme must be prepared in agreement to that stipulated in the standard (ASTM E 185, 2002) which, in turn, refers to the standard (ASTM E 900, 2002) for the prediction of the material embrittlement. For these reasons, Section 4.2.1 is devoted to the first of the procedures mentioned (RG1.99 (2), 1988) while the last one is described in Section 4.2.2. Finally, it is worth noting that alternative models, which can be found in the scientific literature, have been developed: see for example (Odette and Lucas, 1998, 2001; Eason, Wright and Odette, 1998; Yamashita, Iwasaki, Dozaki, 2010); however, these models are considered to be beyond the scope of the present chapter.

#### 4.2.1 The Regulatory Guide Rev. 2

The U.S. Regulatory Guide 1.99, Revision 2 (RG 1.99 (2), 1988) provides the current generic basis to evaluate  $\Delta RT_{NDT}$  (which is considered to be equivalent to  $\Delta T_{41J}$ ) in terms of the copper and nickel contents of the steel and fast neutron fluence (E > 1 MeV) for weld and plate product forms. The model equations for (RG 1.99 (2), 1988) were statistically fitted to a small surveillance database (177 data points) on steels irradiated in surveillance capsules at flux levels somewhat higher than at the vessel wall. The (RG 1.99 (2), 1988) model was derived prior to 1985 and reflects the emerging, but far from complete, physical understanding of embrittlement mechanisms.

The actual (or adjusted) reference temperature  $RT_{NDT}$  for each material in the beltline region is given by expression (6).  $RT_{NDT(U)}$  is the reference temperature for the unirradiated material as defined in (ASME Code). If measured values of initial  $RT_{NDT}$  for the material in question are not available, generic mean values for that class of material may be used if there are sufficient test results to establish a mean and standard deviation for the class. In the case of Linde 80 welds, a generic value  $RT_{NDT(U)}=0^{\circ}F$  is accepted whereas for Linde 0091, 1092, 124 and ARCOS B-5,  $RT_{NDT(U)}=-56^{\circ}F$ . As stated in (RG 1.99 (2), 1988),  $\Delta RT_{NDT}$  must be obtained by applying equation (7); the basis for equation (7) is contained in publications

(Guthrie, 1984) and (Odette et al., 1984). Both contributions used surveillance data from commercial power reactors. The bases for their regression correlations were different in that Odette made greater use of physical models of radiation embrittlement. The two papers contain similar recommendations: (1) separate correlation functions should be used for weld and base metal, (2) the function should be the product of a chemistry factor and a fluence factor, (3) the parameters in the chemistry factor should be the elements copper and nickel, and (4) the fluence factor should provide a trend curve slope of about 0.25 to 0.30 on log-log paper at  $10^{19}$  n/cm<sup>2</sup> (E > 1 MeV), steeper at low fluences and flatter at high fluences.

$$\Delta RT_{NDT} = (CF) \cdot f^{(0.28 - 0.10 \log f)} \tag{7}$$

CF (°F) in equation (7) is the chemistry factor, a function of copper and nickel content. CF is tabulated in (RG 1.99 (2), 1988) for welds and base metal (plates and forgings). Linear interpolation is permitted. If there is no information available, 0.35% copper and 1.0% nickel should be assumed.

The neutron fluence at any depth x in the vessel wall, f(x) (10<sup>19</sup> n/cm<sup>2</sup>, E > 1 MeV), is determined following equation (8):

$$f(x) = f_{SURF} \cdot e^{-0.24 \cdot x} \tag{8}$$

where  $f_{surf}$  (10<sup>19</sup> n/cm<sup>2</sup>, E > 1 MeV) is the calculated value of the neutron fluence at the inner surface of the vessel, and x (in inches) is the depth in the vessel wall measured from the vessel inner surface. Alternatively, if dpa calculations are made as part of the fluence analysis, the ratio of dpa at the depth in question to dpa at the inner surface may be substituted for the exponential attenuation factor in equation (8).

The third term in (6), M, is the quantity, °F, that is to be added to obtain conservative, upperbound values of the adjusted reference temperature. M is obtained through equation (9).

$$M = 2 \cdot \sqrt{\sigma_U^2 + \sigma_\Delta^2} \tag{9}$$

Here,  $\sigma_U$  is the standard deviation for the initial RT<sub>NDT</sub>. If a measured value of initial RT<sub>NDT</sub> for the material in question is available,  $\sigma_U$  is to be estimated from the precision of the test method. If not, generic mean values for that class of material are used. The standard deviation in  $\Delta$ RT<sub>NDT</sub>,  $\sigma_\Delta$ , is 28°F for welds and 17°F for base metal, except that  $\sigma_\Delta$  need not exceed 0.50 times the mean value of  $\Delta$ RT<sub>NDT</sub>.

Finally, when two or more credible surveillance data sets become available from the reactor in question, they may be used to determine the adjusted reference temperature of the beltline materials. In this case, if there is clear evidence that the copper or nickel content of the surveillance weld differs from that of the vessel weld, the measured values of  $\Delta RT_{NDT}$ should be adjusted by multiplying them by the ratio of the chemistry factor for the vessel weld, (CF)<sub>V</sub>, to that of the surveillance capsule weld, (CF)<sub>C</sub>, see equation (10).

$$\left(\Delta RT_{NDT}\right)_{V} = \left(\Delta RT_{NDT}\right)_{C} \cdot \frac{\left(CF\right)_{V}}{\left(CF\right)_{C}}$$
(10)

Second, the surveillance data should be fitted using equation (7) to obtain the relationship of  $\Delta RT_{NDT}$  to fluence. To do so, calculate the chemistry factor, CF, for the best fit by

multiplying each adjusted  $\Delta RT_{NDT}$  by its corresponding fluence factor, summing the products, and dividing by the sum of the squares of the fluence factors. The resulting value of CF will give the relationship of  $\Delta RT_{NDT}$  to fluence that fits the plant surveillance data in such a way as to minimise the sum of the squares of the errors.

#### 4.2.2 Standard ASTM E 900 - 02

The purpose of ASTM Standard E 900-02 (ASTM E 900, 2002) is to establish an improved correlation in respect of that proposed by the Regulatory Guide (RG1.99 (2), 1988) which allowed the transition temperature shift,  $\Delta T_{41J}$ , to be obtained as a function of the neutron fluence. The data base used in this case consisted of 600 data; thus, it is much more robust than that of the Regulatory Guide where only 177 data were available. Here, the expressions are not purely phenomenological but physically guided, taking into consideration when possible the nanofeatures participating in the process (described above). Another interesting feature is that the ASTM E 900-02 formulation is based on robust statistical techniques for the treatment of the large data set (non-linear, least-square regression analysis). It is worth noting that the origin of this procedure relies on the expressions proposed by (Eason et al., 1998). The procedure distinguishes between the three mechanisms described in Section 4.1:

- Stable matrix damage (SMD) associated with the presence of point defects and loop dislocations.
- Copper rich precipitates (CRPs).
- Grain Boundary Segregations (GBSs) of embrittling elements as phosphorus.

As there is no evidence of the influence of this last mechanism on USA vessels, the form of the correlation involves only the two major embrittlement terms: the SMD and the CRP; nevertheless, the influence of phosphorus is indirectly present through the SMD. The mean value of the transition temperature shift is calculated as follows (11):

$$Shift = SMD + CRP \tag{11}$$

The formulas for both terms must take into consideration the empirical reality; specifically, the CRP mechanism saturates with fluence while, on the contrary, the SMD damage process grows monotonically, apparently without limit.

Expression (12) represents the transition temperature shift, in °F, due to the SMD

$$SMD = 6.70 \cdot 10^{-18} \cdot e^{\frac{20730}{T_i + 460}} \cdot (\Phi)^{0.5076}$$
(12)

The main characteristics as well as the meaning of the variables are explained below:

- The influence of irradiation temperature, T<sub>i</sub>, in °F, in the range 275-295 °C (527 563 °F, respectively) is modelled by means of an exponential function.
- The effect of the irradiation, which is expressed in the neutron fluence,  $\Phi$  (n cm<sup>-2</sup>, E > 1 MeV), increases indefinitely, without saturation.
- There is no explicit dependence on the neutron flux,  $\varphi$ .

The second term in (11) which represents the shift in the transition temperature due to the CRP mechanism, in °F, responds to formula (13), together with expressions (14) and (15):

$$CRP = B \cdot (1 + 2.106 \cdot Ni^{1.173}) \cdot F(Cu) \cdot G(\Phi)$$
(13)

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$$F(Cu) = (Cu - 0.072)^{0.577}$$
(14)

where Cu≤0.305 in general and Cu≤0.25 for Linde 80 and 0091.

$$G(\Phi) = \frac{1}{2} + \frac{1}{2} \cdot Tanh\left[\frac{\log(\Phi) - 18.24}{1.052}\right]$$
(15)

The relevant features in (13-15) and the meaning of the variables is explained in the following:

- The coefficient B takes different values depending on the material (B=234, for welds; B=128 for forgings; B=208 for combustion engineering plates; B=156 for other plates).
- There is a strong influence of the nickel content, Ni (expressed in % wt.).
- The influence of Cu is taken into consideration through F(Cu) (14); this effect occurs only for Cu>0.072% and saturates for Cu>0.305% (0.25% for Linde 80 and 0091)
- The irradiation temperature is not considered to play a role.
- The neutron fluence  $\Phi$  influence is represented through the term  $G(\Phi)$  where the saturation is modelled as a hyperbolic tangent function.

Moreover, a term corresponding to the standard deviation  $\sigma_{TTS}=22^{\circ}F$  must be considered. It takes into consideration the uncertainties in the input data and in the preparation of the model.

# 5. The Master Curve model for the description of the fracture toughness in the DBT region

#### 5.1 Description of the model

Advances in fracture mechanics technology have made it possible to improve the semiempirical indirect methodology described above, currently in force to describe the fracture toughness in the DBT region in different aspects. First, the development of EPFM allows fracture toughness values to be determined using much smaller specimens and utilising J integral techniques, that is, measuring values of K<sub>Jc</sub> instead of K<sub>Ic</sub>. Moreover, the analytical techniques for structural integrity assessment can now be expressed in terms of EPFM. The first issue, which is considered relevant for the contents of this chapter, is described in the present section; indeed, the scope is explicitly focused on the Master Curve (MC) approach to describe the fracture toughness of vessel steels in the DBT region.

The MC model, originally proposed by Wallin (Wallin, 1984; Wallin et al. 1984; Wallin, 1989; Wallin, 1995), provides a reliable tool based on a direct characterisation of the fracture toughness in the DBT region. This approach is a consequence of the developments in EPFM together with an increased understanding of the micro-mechanisms of cleavage fracture. Valiente et al. (Valiente et al., 2005) have briefly but comprehensively reviewed the previous contributions made to understand cleavage in a ferritic matrix that leads to the MC approach. The basic MC method for analysis of brittle fracture test results is defined in the standard ASTM E 1921 (ASTM E 1921, 2009). The mathematical and empirical details of the procedure are available in (Merkle et al., 1998; Ferreño, 2008; Ferreño et al. 2009). The main features and advantages of the method are hereafter summarised:

• MC assumes that cleavage fracture in non austenitic steels is triggered by the presence of particles close to the crack tip. Therefore, fracture is mainly an initiation dependent process. As a consequence, fracture is governed by weakest link statistics which follows a three parameter Weibull distribution. For small-scale yielding conditions, therefore using EPFM, the cumulative failure probability, P<sub>f</sub>, is given by equation (16):

$$P_f = 1 - e^{-\frac{B}{B_0} \cdot \left(\frac{K_{lc} - K_{\min}}{K_0 - K_{\min}}\right)^m}$$
(16)

where  $K_{Jc}$  is the fracture toughness for the selected failure probability,  $P_f$ ,  $K_0$  is a characteristic fracture toughness corresponding to 63.2% cumulative failure probability, B is the specimen thickness and  $B_0$  a reference specimen thickness,  $B_0 = 25.4$  mm. The experimental data allows the Weibull exponent, m=4, to be fixed (Merkle et al., 1998) and the minimum value of fracture toughness for the probability density function,  $K_{min} = 20$  MPa m<sup>1/2</sup>. Therefore, only  $K_0$  must be estimated from the empirical available data.

• The dependence between K<sub>0</sub> (in MPa m<sup>1/2</sup>) and temperature (°C) for cleavage fracture toughness is assumed to be (17):

$$K_0 = 31 + 77 \cdot e^{0.019 \cdot (T - T_0)} \tag{17}$$

where  $T_0$  is the so-called MC reference temperature; it corresponds to the temperature where the median fracture toughness for a 25 mm thickness specimen (1T, according to ASTM terminology) has the value 100 MPa m<sup>1/2</sup>.

• One of the main advantages of the method is that it allows data from different size specimens to be compared. As thickness increases, the toughness is reduced, due to the higher probability of finding a critical particle for the applied load. The ASTM standard (ASTM E 1921, 2009) provides expressions to relate the fracture toughness for specimens of different thicknesses. Equation (16) can be re-written considering the same failure cumulative probability, P<sub>f</sub>, for two specimens of different thickness, namely B<sub>1</sub> and B<sub>2</sub>, thus leading to expression (18):

$$K_{J_c,2} = K_{\min} + \left(K_{J_c,1} - K_{\min}\right) \cdot \left(\frac{B_2}{B_1}\right)^{\frac{1}{4}}$$
(18)

• The distribution fitting procedure involves finding the optimum value of T<sub>0</sub> for a particular set of data. For this task, all data are thickness adjusted to the reference specimen thickness B<sub>0</sub> = 25.4 mm using equation (18). The procedure can be applied either to a single test temperature or to a transition curve data, T<sub>i</sub> being the generic temperature of the different tests. In the latter approach (the former is just a particular case) T<sub>0</sub> is estimated from the size adjusted K<sub>JC</sub> data (K<sub>JC,1T</sub>) using a multi-temperature maximum likelihood expression (see equation (19)). To estimate the reference temperature, T<sub>0</sub>, a previous censoring of the non size-adjusted data must be applied. Fracture toughness data that are greater than the validity limit given by equation (20), as defined in (ASTM E 1921, 2009), are reduced to the validity limit, K<sub>Jc(lim)</sub> and treated

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as censored values in the subsequent estimation stage ( $\delta_i = 0$  in expression (19)). This condition is imposed to guarantee high constraint conditions in the crack front during the fracture process.

$$\sum_{i=1}^{N} \delta_{i} \cdot \frac{e^{0.019 \cdot (T_{i} - T_{0})}}{11 + 77 \cdot e^{0.019 \cdot (T_{i} - T_{0})}} - \sum_{i=1}^{N} \frac{\left(K_{Jc,i} - 20\right)^{4} \cdot e^{0.019 \cdot (T_{i} - T_{0})}}{\left[11 + 77 \cdot e^{0.019 \cdot (T_{i} - T_{0})}\right]^{5}} = 0$$
(19)  
$$K_{Jc(\text{lim})} = \sqrt{\frac{E \cdot \sigma_{Y} \cdot b_{0}}{30 \cdot \left(1 - \nu^{2}\right)}}$$
(20)

In equation (20)  $\sigma_y$  is the yield strength at test temperature, E is the Young's modulus,  $b_0$  the initial ligament and v is the Poisson's ratio. It must be stressed that factor 30 in equation (20) is currently under discussion (Merkle et al., 1998) and that, for instance, the ASTM E1820-01 Standard (ASTM E 1820, 2001) imposes a more demanding limit with a factor 50 or 100 depending on the nature of the steel.

• The standard deviation in the estimate of T<sub>0</sub>, expressed in °C, is given by (21):

$$\sigma_{T_0} = \frac{\beta}{\sqrt{r}} \tag{21}$$

where r represents the total number of valid specimens (not censored results) used to establish T<sub>0</sub>. The values of the factor  $\beta$  are provided in (ASTM E 1921, 2009).

- The statistical analysis can be reliably performed even with a small number of fracture toughness tests (usually between 6 and 10 specimens). Moreover, as an EPFM approach is used, the specimen size requirement, equation (20), is much less demanding than that of the LEFM (ASTM E 399, 2009). These remarks are of great relevance in nuclear reactor surveillance programmes where the amount of material available is usually very limited and consists of small size samples (Charpy specimens).
- By rearranging equations (16) and (17) it is possible to obtain expression (22) which provides an estimate of  $K_{Jc}$  for a given cumulative failure probability,  $P_f$ , once  $T_0$  has been determined. In this way, the confidence bounds of the distribution (usually taking  $P_f = 0.01$  or 0.05 for the lower bound and 0.95 or 0.99 for the upper bound) can be obtained. As a particular case, the expression for the median fracture toughness ( $P_f = 0.5$ ) (see Equation (23)) is determined.

$$K_{J_c,P_f} = K_{\min} + \left[ -\ln\left(1 - P_f\right) \right]^{0.25} \cdot \left[ 11 + 77 \cdot e^{0.019 \cdot (T - T_0)} \right]$$
(22)

$$K_{J_c(med)} = 30 + 70 \cdot e^{0.019(T - T_0)}$$
(23)

• Finally, any test that does not fulfil the requirement for crack front straightness or that terminates in cleavage after more than a limit of slow-stable crack growth will also be regarded as invalid.

#### 5.2 Open issues concerning the Master Curve approach

The MC has become a mature tool for characterising the fracture toughness of ferritic steels in the DBT region. Considerable empirical evidence provides testament to the robustness of the MC procedure. One of the main advantages of this procedure relies on the possibility of assessing the state of a RPV vessel by direct measurement of fracture toughness rather than through the use of the currently accepted correlative approaches, based on Charpy tests. The procedure currently accepted to assess the steel neutron embrittlement partially incorporates the MC reference temperature concept, T<sub>0</sub>; in this sense, to enable the use of the MC methodology without completely modifying the structure of the ASME code the approach stated in code cases N-629 (ASME CC 629, 1999) and N-631 (ASME CC 631, 1999) was adopted. It consists of defining a new index temperature, RT<sub>T0</sub>, for the K<sub>Ic</sub> and K<sub>IR</sub> ASME curves (4, 5), as an alternative to  $RT_{NDT}$ . The definition of  $RT_{T0}$  is given in equation (24). This value of  $RT_{T0}$  is set, see (VanDerSluys et al., 2001), by imposing that the ASME K<sub>Ic</sub> curve indexed with RT<sub>T0</sub> in place of RT<sub>NDT</sub> will bound the majority of the actual material fracture toughness data. In this sense, RT<sub>T0</sub> was set such that the corresponding ASME K<sub>Ic</sub> curve falls below the MC 95% confidence bound for at least 95% of the data generated with 1T specimens.

$$RT_{T_0} = T_0 + 19.4 \,^{\circ}C \tag{24}$$

Evidently, this approach, currently in force, is merely a compromise solution that attempts to fit the new concepts into the old structure. Apart from this practical aspect, several other open issues remain concerning the application of the MC as well as theoretical aspects. The following issues must be emphasised:

- There is no experimental data that allows the MC to be used in special applications such as irradiated materials with high neutron fluence, materials susceptible to intergranular fracture or materials showing exceptional lower-shelf or transition behaviour. Indeed, the main feature of the MC method consisting of assuming that the dependence of the fracture toughness of a material on temperature in the transition range is not sensitive to characteristics such as the mechanical properties and the microstructure is purely speculative for the cases mentioned above.
- The published literature shows that the PCCv (Pre Cracked Charpy-V Notched) specimen analysed using the ASTM Standard E 1921 (ASTM E 1921, 2009) generally shows a reference temperature ~10 °C lower than the CT (compact tension) specimen. Compared with the inherent scatter in the transition temperature, this difference is small. However, it has been observed in many materials. Although different hypotheses were proposed a decade ago in order to explain this fact, the current consensus in the scientific community is that this difference is motivated by the different level of constraint in single edge notch bend and CT geometries.
- The above issue is a particular case of the general question of how crack-tip constraint effects (stress tri-axiality in the vicinity of the crack tip) can be described. In fracture mechanics, it is well known that crack-tip constraint can be influenced by loading (out of plane or multi-axial loading) or by the crack shape and crack depth to ligament ratio. Nevertheless, to date, there is no agreement about how to manage crack tip constraint in the practical application of structures and components containing postulated or real cracks and made of ferritic steel.

• The MC approach procedure standardised in ASTM E1921 (ASTM E 1921, 2009) is defined for quasi-static loading conditions. However, the extension of the MC method to dynamic testing is still under discussion although a great effort has been made over the last decade to qualify the method for dynamic loading conditions and to use it for structural purposes.

This list of questions currently under discussion reveals that, although the MC methodology is increasingly being recognised as an attractive alternative for describing the fracture toughness of ferritic steels in the DBT region, further research needs to be done in order to properly deal with the open issues mentioned above.

### 6. Conclusions

The purpose of this chapter was to provide the readers with an introductory text, selfcontained insofar as possible, concerning the current state of the art in the process of embrittlement that takes place in nuclear vessel steels, paying particular attention to the ductile to brittle transition region. It was the purpose of the authors to introduce the topics in a logical sequence in an attempt to explain the scientific and historical arguments that justify the different methods and tools currently available. A phenomenological and scientific description of the causes and consequences of material embrittlement was presented. An explanatory description of the characterisation tools that are available for the nuclear facilities -implemented in their surveillance programmes- to determine the evolution of the fracture toughness of the vessel steel throughout the operative lifetime of the plant, emphasising their advantages and limitations, was also included. This leads, in a natural way, to the Master Curve methodology, as an alternative procedure for obtaining, in the context of Elastic-Plastic Fracture Mechanics, the material fracture toughness; as stressed in the text, this procedure offers many advantages and few limitations, which is why it is widely used at present in a great number of ambitious scientific research projects. It is the opinion of the authors that all of the evidence available points to the fact that the Master Curve approach is set to become an indispensable ingredient in the future of surveillance programmes.

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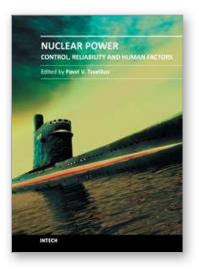
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