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Journal of Nuclear Materials 367–370 (2007) 575–580

journal of nuclear materials

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A closer look at the fracture toughness of ferritic/martensitic steels

Enrico Lucon *

SCK·CEN, Institute for Nuclear Materials Science, Boeretang 200, B-2400 Mol, Belgium

Abstract

SCK·CEN has characterized the mechanical properties of several ferritic/martensitic steels, both unirradiated and irradiated. Fracture toughness has been evaluated using Charpy impact and fracture mechanics tests. Two safety-related features have emerged: (a) the applicability of the master curve approach (ASTM E1921-05) appears questionable; and (b) irradiation embrittlement is systematically larger when quantified in terms of quasi-static fracture toughness than when measured from Charpy tests. Both issues are examined in detail and possible interpretations are proposed; potential improvements given by the application of more advanced fracture toughness analysis methodologies are discussed. In order to clarify whether the Charpy/fracture toughness difference in embrittlement is due to loading rate effects, dynamic toughness tests have been performed in the unirradiated condition and for two irradiation doses (0.3 and 1.6 dpa). The corresponding dynamic T_0 shifts have been compared with the shifts of Charpy and master curve quasi-static transition temperatures. Other possible contributions are examined and discussed.

1. Introduction

Ferritic/martensitic (F/M) steels, with chromium contents between 9% and 12%, have been considered as structural materials for fusion reactors since the late 1970s [1,2], after having been previously considered in fast reactor programmes [1,3]. In the mid 1980s, the concept of low-activation materials (later to be revised as reduced-activation) was introduced into international fusion programs [4,5].

More recently, a new generation (Generation IV) of fission reactors has been proposed, which are expected to produce energy under safe and proliferation-resistant conditions [6]; their service conditions also envisage the use of F/M steels as possible structural and cladding materials [7]. The same materials are also primary structural candidates for accelerator driven systems (ADS) [8].

For any structural material, the characterization of the main mechanical properties (tensile, fracture toughness, fatigue, creep, etc.) and their evolution as a consequence of irradiation exposure is of paramount importance. More specifically, for components expected to operate at relatively low temperatures ($T < 400$ °C) during at least part of their service life, fracture toughness has to be assessed as well as possible embrittlement phenomena which could occur during operation.

The conventional approach to investigating irradiation embrittlement is by means of Charpy impact

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* Tel.: +32 14 333088; fax: +32 14 321216.
E-mail address: elucon@sckcen.be

0022-3115/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2007.03.037

tests, using the DBTT (ductile-to-brittle transition temperature) and its irradiation-induced shift as reference parameters. Moreover, particularly in the fusion community, the use of sub-size (KLST) rather than full-size Charpy specimens is quite common.

More and more often, however, investigators resort to the direct measurement of fracture toughness using fatigue precracked specimens. When samples are tested within the ductile-to-brittle transition region, a relatively limited number of tests is sufficient to establish the so-called reference temperature (T_0) using the well-established master curve (MC) methodology [9,10]. Embrittlement effects can therefore be assessed in terms of an increase in the reference temperature (ΔT_0) rather than in the Charpy-based ΔDBTT.

In recent years, SCK·CEN has extensively characterized by means of Charpy and fracture toughness tests the toughness properties of several F/M steels [11,12] in unirradiated and irradiated conditions: EUROFER97 (E97, European reference structural steel for fusion applications), EM10 (9Cr1Mo), T91 (Mod 9Cr–1Mo) and HT9 (12Cr–1MoWV). More details on the individual materials and on the test procedures can be found in [11,12].

From the investigations performed, two distinct features have emerged, both bearing potentially serious safety implications:

1. the dubious applicability of the MC methodology to F/M steels, which has been already questioned by some investigators [13], and more specifically the apparent inability of the method to fully account for the significant scatter in the fracture toughness results; and
2. the systematic underestimation of embrittlement effects when using Charpy-based DBTT shifts, rather than reference temperature shifts [14].

Possible interpretations of both issues are proposed and discussed in detail within this paper. A general framework for the topics addressed in this paper is also given in [15].

2. Applicability of the master curve method to F/M steels

The MC approach, based on the weakest-link theory [16] applied to a three-parameter Weibull distribution of fracture toughness values in the transition range, is nowadays widely used for treating statistical size effects in cleavage fracture. Statistical methods are employed to predict the transition toughness curve and specified tolerance bounds for standard-size specimens of the material tested.

This method is nominally restricted to macroscopically homogeneous ferritic steels with yield strengths in the range 275–825 MPa and weld metals having less than 10% strength mismatch with respect to the base metal. Nonuniform or inhomogeneous materials, such as multipass weldments, are in principle not amenable to this analytical treatment.

Fig. 1 shows a normalized plot of the 160 fracture toughness test results obtained at SCK·CEN on E97, EM10, T91 and HT9, in the unirradiated and irradiated (different doses and irradiation temperatures) conditions. If we only consider the applicability range of the MC approach ($-50\,^{\circ}\mathrm{C} \leqslant T - T_0 \leqslant 50\,^{\circ}\mathrm{C}$) and concentrate our attention on the data points falling below the 5% and 1% lower bounds, we observe that the corresponding percentages (13.3% and 4.2%) are significantly higher than the predictions of the method.

For the overall data set shown in Fig. 1, the value of the Weibull exponent m has been evaluated using the Generalized Maximum Likelihood (GML) method, which also allows estimating the standard deviation σ_{m} as a function of the number of valid data [17]. The results obtained for the $\pm 3\sigma$ ($\pm$99%) confidence interval indicate: $m = 1.69$–3.50, which does not include the theoretical value of the Weibull exponent according to the MC approach ($m = 4$). Note also that the lower is m, the higher is the material's scatter (or inhomogeneity); moreover, the more pronounced is constraint loss [18].

Both experimental and analytical considerations, therefore, seem to raise doubts about the full applicability of the MC to F/M steels and call for the use of alternative approaches. Nowadays, the following more advanced analytical approaches are available for the fracture toughness assessment of inhomogeneous materials [19]:

(a) The SINTAP lower tail (SLT) analysis procedure [20], which is very effective in providing realistic lower-bound type estimates even for highly inhomogeneous materials, although it cannot describe the whole distribution.
(b) The single point estimation (SPE) method [19], which delivers a rather crude but effective engineering assessment of significantly inho-

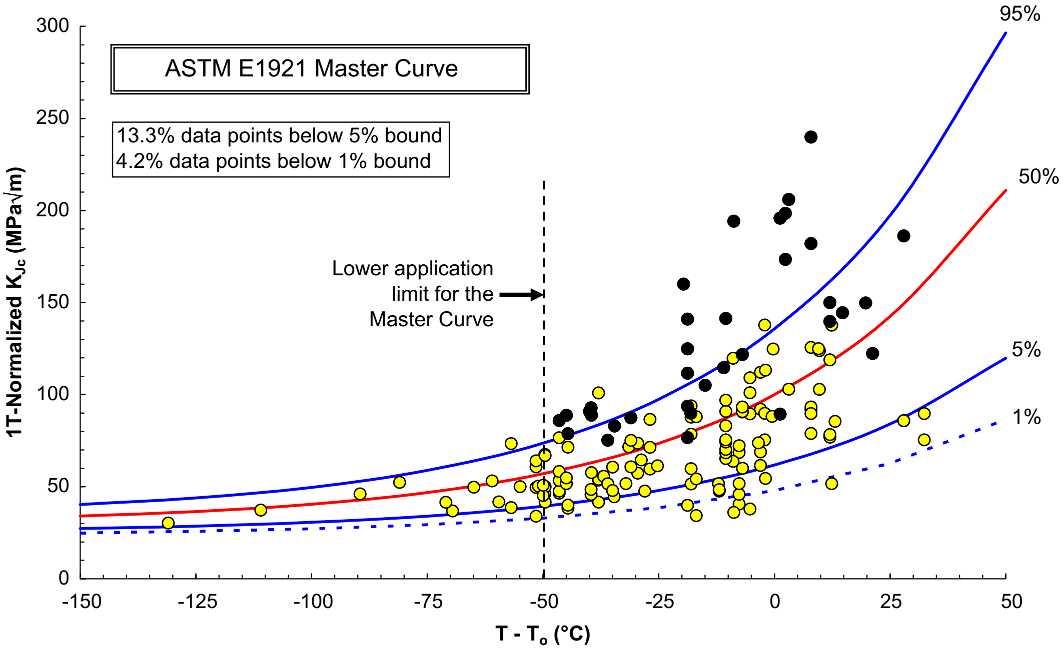

Fig. 1. MC analysis of 160 fracture toughness test results on E97, EM10, T91 and HT9. Black solid points are invalid data according to E1921-05.

mogeneous data sets, based on the derivation of individual T_0 estimates from all non-censored values.

(c) The multi-modal master curve (MMMC) [19], which relies on the maximum likelihood estimate for random inhomogeneity and on the comparison between experimental and theoretical scatter.

The SPE and MMMC methods are particularly suited to the analysis of data sets including different materials.

The three methods have been applied to the fracture toughness data set shown in Fig. 1. While no significant changes have been observed in terms of individual T_0 or ΔT_0 (irradiation shift) values, the use of the SPE and MMMC methods has proven

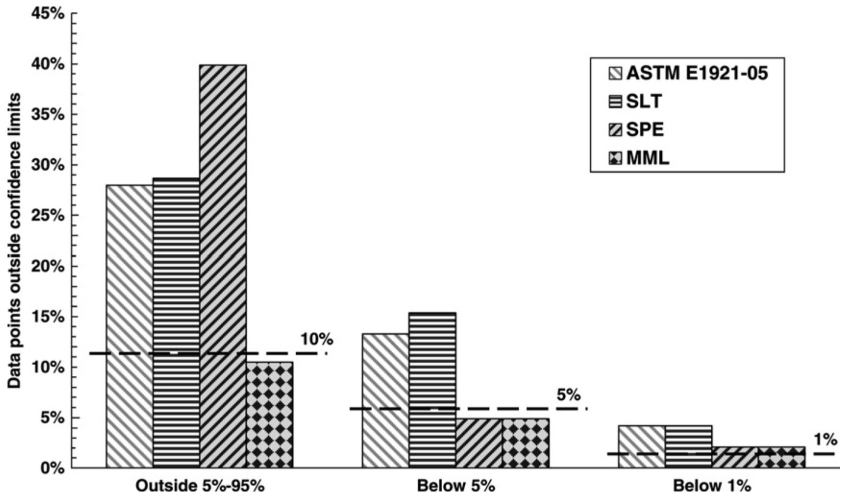

Fig. 2. Application of advanced analytical approaches to the fracture toughness test results of F/M steels.

to be effective in reducing the occurrence of low-toughness outliers to acceptable levels (for both methods, 4.9% below the 5% limit and 2.1% below the 1% limit). The MMMC method, however, provides a better representation of the overall distribution, considering also high-toughness outliers (10.5% outside the 5–95% confidence bounds, as compared to 28% for the conventional MC). Using the SLT approach does not yield significant improvements (15.4% below 5% and 4.2% below 1%; 28.7% outside 5–95%). The results of the analyses performed are shown in Fig. 2.

It is also worth mentioning that, according to the MMMC approach, only two of the thirteen investigated fracture toughness data sets can be classified as originating from a homogeneous material.

3. Discrepancies between Charpy and fracture toughness measurements of irradiation embrittlement

As noted above, a systematic and significant underestimation of irradiation embrittlement has been noted when Charpy shifts (ΔDBTT) are compared to direct fracture toughness (ΔT_0) results for several F/M steels tested at SCK·CEN before and after irradiation. Similar results have been reported in the literature [14,21,22]. This is clearly shown in Fig. 3, which also includes similar information for a range of low-alloy ferritic reactor pressure vessel (RPV) steels irradiated to much lower doses. The irradiation shifts measured from Charpy tests are in substantial agreement with reference temperature shifts obtained from fracture toughness tests for RPV steels.

An even larger discrepancy in terms of upper shelf fracture toughness behaviour has been recently reported [23] for EUROFER97 irradiated at 300 °C up to 1.6 dpa; a drop of 81.4% in J-integral ductile fracture initiation (J_Q) corresponds to only 7.5% decrease in Charpy upper shelf energy (USE).

Two distinct arguments are proposed to explain the observed effects; they are examined in detail in the following sections.

3.1. Loading rate effects

The most straightforward difference between Charpy impact and quasi-static fracture toughness tests, besides the absence or presence of a crack in the specimen, is the loading rate, which differs by several orders of magnitude. In order to assess the influence of loading rate on embrittlement shifts in F/M steels, dynamic (impact) fracture toughness tests have been performed on precracked Charpy specimens of EUROFER97 in the unirradiated condition and for two irradiation doses (0.3 and 1.6 dpa). Tests have been performed using an instrumented impact pendulum with striker conforming to the ISO 148 requirements (2 mm tup radius) at impact rates between 1 and 1.5 m/s; results have been analyzed using the master curve approach.

For the investigated conditions, the shifts of the dynamic reference temperature ($\Delta T_{0,\mathrm{dyn}}$) fall between the results of the Charpy and the quasi-static toughness tests, showing that loading rate effects can only partially explain the observed differences.

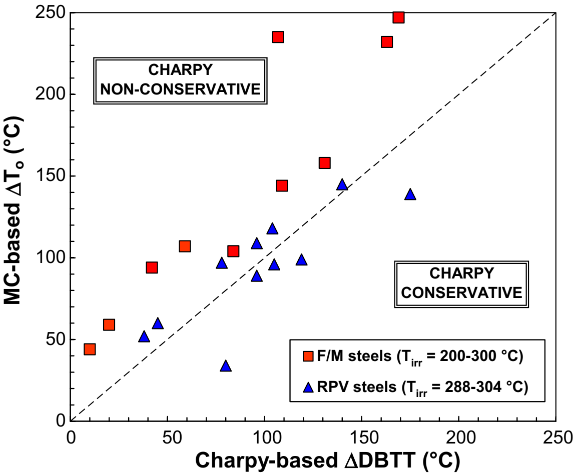

Fig. 3. Comparison between irradiation shifts measured from Charpy and fracture toughness tests for F/M and RPV steels.

3.2. Intrinsic nature of the parameters used to index transition fracture behaviour: initiation (T_0) versus initiation + propagation (DBTT)

When irradiation embrittlement effects are quantified by the combined use of DBTT from Charpy tests and T_0 from fracture toughness tests, the different intrinsic nature of these two transition temperatures has to be accounted for. Typically, the DBTT corresponds to the point equidistant between lower shelf (fully brittle) and upper shelf (fully ductile) conditions, where the material has undergone not only fracture initiation but also a considerable degree of ductile crack propagation and gross plasticity. On the other hand, the reference temperature T_0 is defined as the temperature at which the median toughness K_{Jc} of a standard 1-in thick specimen is

100 MPa$\sqrt{m}$, which for most materials corresponds to almost pure cleavage conditions (no stable propagation – just initiation and unstable fracture). Indeed, the MC method invalidates test results which exceed the measuring capacity of the specimen or when stable crack growth exceeds 5% of the initial uncracked ligament.

More specifically, for unirradiated and irradiated (0.3, 0.7 and 1.6 dpa) E97, the Charpy DBTT temperatures correspond to median toughness values ranging from 155 to 284 MPa$\sqrt{m}$. Conversely, T_0 on the Charpy curve always corresponds to full lower shelf conditions (less than 1% ductile fracture appearance on the fracture surface).

Therefore, the much higher increases of T_0 observed for F/M steels could be related to a much larger effect of irradiation on fracture initiation than on fracture propagation, coupled with the detrimental effect of slower (quasi-static) loading rates. To demonstrate this point, an alternative Charpy index temperature can be considered: T_I, which corresponds to the onset of ductility on the Charpy transition curve, and is therefore strictly related to the initiation of cleavage fracture and bears no contribution from the propagation stage. It can also be defined as the temperature above which the force at general yield (F_{gy}) and the maximum force (F_m) start to deviate from each other. This parameter is evaluated in the framework of the so-called 'load diagram approach' [24,25], which allows deriving more fundamental information from the analysis of the force/deflection traces of instrumented Charpy tests. If ΔT_I shifts are evaluated from the instrumented Charpy tests performed on unirradiated and irradiated E97, the corresponding shifts are in much closer agreement with ΔT_0 (initiation only) than ΔDBTT (initiation + propagation). The small remaining difference can most likely be attributed to loading rate effects.

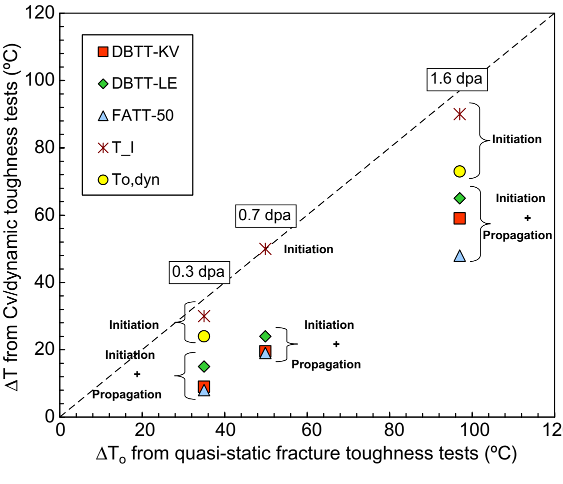

Fig. 4. Role of initiation and propagation of fracture in irradiation shifts measured from Charpy and fracture toughness tests on F/M steels.

The overall situation for E97 in the different conditions is clearly depicted in Fig. 4, where the results of the previously mentioned dynamic toughness results are also included. In the figure, three different Charpy-based transition temperatures are reported (DBTT-KV = from the absorbed energy/temperature curve; DBTT-LE = from the lateral expansion/temperature curve; FATT-50 = from the shear fracture appearance/temperature curve).

4. Conclusions

The results of 160 fracture toughness tests performed on four different F/M steels (E97, EM10, T91 and HT9) in unirradiated and various irradiated conditions have been analyzed in detail. The most relevant conclusions are the following:

1. The conventional master curve methodology appears unable to fully account for the apparent inhomogeneity of the steels. More specifically, the statistical lower bounds (5% and 1%) do not account for the presence of a significant amount of low-fracture toughness results, which represents a serious concern for the safety assessment of structural materials. The use of advanced analytical approaches, such as the single point estimation method or the multi-modal master curve, appears clearly beneficial in this respect.
2. Assessing neutron embrittlement by means of traditional Charpy tests can lead to serious underestimation of the actual degradation of the materials' toughness. The differences observed between Charpy and fracture toughness temperature shifts can be explained in terms of two effects:
   (1) influence of lower loading rates;
   (2) much more pronounced degradation of toughness in terms of fracture initiation (indexed by the master curve reference temperature T_0) than fracture propagation (associated with the conventional Charpy transition temperature).

Based on the investigations performed, characterizing the actual fracture toughness properties of

F/M steels is recommended, rather than relying only on Charpy data, for the assessment of irradiation-induced embrittlement. The use of the multi-modal master curve approach is also advised for the analysis of fracture toughness test results, on account of the better representation of the apparent inhomogeneity of F/M steels.

Additional issues which could be of relevance, although they have not been addressed in this paper, include loss of constraint which is quite significant for RAFM steels (particularly after low temperature and high dose irradiation) and the constraint limit $M = 30$ given in the current version of the MC, which has been contended to be too lenient.

Acknowledgements

The Author is gratefully indebted to the technical staff of the RMO department of SCK·CEN. My gratitude also to R. Chaouadi and M. Scibetta for the many fruitful discussions. This work was partially accomplished within one of the tasks of the EFDA Long-Term Programme.

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