Application of local approach to fracture of an RPV steel: effect of the crystal plasticity on the critical carbide size.

Pierre Forget*,a, Bernard Marini*a, Ludovic Vincenta

aCEA, DEN-Service de Recherches Métallurgiques Appliquées, Université Paris-Saclay, Gif-sur-Yvette, F-91191, France

Abstract

Avoidance of brittle fracture is a key issue of the integrity assessment of the nuclear reactor pressure vessels (RPV). The microstructural heterogeneities of these heavy components are in direct relation with the scatter of brittle fracture toughness. A microstructure informed brittle fracture (MIBF) model is used here to capture the effect of some microstructural features. This local approach type model is based on the Griffith criterion for brittle fracture applied to micro cracks nucleated on carbides. The size distribution of the cleavage initiators introduces a source of scatter for the fracture stress. The originality of the MIBF model is to introduce a second source of scatter: the stress distribution inside a representative aggregate of the bainitic microstructure. These stresses are the average for each bainitic packet of the maximum principal stresses. Their non-uniform distribution is due to the incompatibilities of plastic deformation introduced by crystallographic misorientations between packets. Finite elements simulations on aggregates representative of the steel bainitic microstructure are performed to identify the stress distributions. The size distribution of cleavage initiators is the carbide size distribution as the carbides cracking induced by plastic deformation is considered to be the main damage leading to cleavage propagation in RPV steel. The only free parameter of the model is the effective surface energy γf. Applied to the Euro fracture toughness dataset, the MIBF model predicts the specimen size effect on fracture toughness as well as the temperature dependence of the fracture toughness up to 200 MPa√m. The analysis of the numerical results shows that taking into account the incompatibility stresses widens the size range of carbides potentially implied in the fracture process. However, approximatively half of these carbides have still a size larger than the largest observed sizes underlining the importance to perform the determination of size distributions on a very large number of precipitates.

© 2016 The Authors. Published by Elsevier B.V.
Peer-review under responsibility of the Scientific Committee of ECF21.

Keywords: fracture toughness ; brittle fracture; RPV steel ; microstructure; carbides ; local approach ; MIBF model

* Corresponding author. Tel.: +33-1-69087896; fax: +33-169087167.
E-mail address: pierre.forget@cea.fr
1. Introduction

Brittle fracture prediction plays an important role for the integrity assessment of the nuclear reactor pressure vessels (RPV). In these heavy components, heterogeneities of the steel microstructure are induced by the fabrication process and are an important source of scatter of the fracture toughness. In order to identify the features controlling this scatter, a microstructure informed brittle fracture (MIBF) model is used to capture the effects of some microstructure characteristics i.e. the scatter of the critical stress induced by the carbide size distribution as well as the incompatibility stresses arising from the poly-crystalline structure under plastic deformation. This model is developed in the frame of the Local Approach to Fracture (LAF) defined by Beremin (1983) and used since then to predict the brittle fracture of low alloy steels (Pineau 2006). At CEA this approach has been continuously developed over this period: Kantidis (1992) applied the Beremin model to intergranular fracture, Renevey (1996) developed a model taking into account the effect of large manganese sulfide clusters, Carassou (1998) used this model in conjunction with a ductile tearing model to describe the ductile to brittle transition, and Forget (2001) and Parrot (2006) used the Beremin model for Charpy modelling. In all these applications, it was necessary to introduce a dependency of Beremin critical stress $\sigma_u$ with the temperature and neutron fluence to predict both the brittle-ductile toughness transition curve and its evolution with irradiation. The so-called MIBF model was developed in this context to propose an alternative to such phenomenological modifications, by incorporating additional physical effects at the microstructure scale.

2. Brittle fracture model

The basic idea of the MIBF model is to express the variability of brittle fracture in a LAF model not only in terms of the distribution of defects but also as a function of the inherent stress heterogeneity that prevails around these defects due to local strain incompatibilities at the aggregate scale.

![Fig. 1. The three scales of the MIBF model from left to right: the fracture toughness specimen composed of elementary volume $V_0$, the volume $V_0$ containing several bainitic packets and the packet of volume $v$ containing several carbides.](image)

Let’s suppose that a loading $P$ is applied to the specimen (Fig. 1). The specimen is composed of elementary volumes $V_0$ which are subject to a macroscopic stress field $\Sigma$. $\Sigma$ is the maximum principal stress applied on the boundaries of the elementary volume $V_0$. This quantity is constant in $V_0$ for the classical LAF models. Here, the maximum principal stress inside $V_0$, named $\sigma^*$, is not uniform and is distributed around $\Sigma$ according to a distribution function $P(\sigma^* > \sigma)$. $\sigma^*$ is considered as the driving force at the scale of bainitic packets for the propagation of cracks initiated on carbides. It is hence an average of the maximum principal stress over the volume $v$ of each bainitic packet. The distribution of such quantity can be identified on the results of Finite Element simulations performed at the polycrystal level, provided the crystal plasticity law used in the simulations can reproduce some essential physical features of the plastic behaviour of the ferritic steel, such as the different slip systems of Fe-BCC and their thermal activation for instance.

Considering that each cracked carbide inside a bainitic packet behaves as a Griffith crack, failure occurs when $\sigma^*$ overcomes a critical value $\sigma_c$: 
\[
\sigma_c = \sqrt{\pi \frac{E \cdot \gamma_f}{2(1-\nu^2)}} \frac{r}{\gamma_i}
\]
where \( r \) is the radius of the broken carbide, \( E \) is Young’s modulus, \( \nu \) is Poisson coefficient and \( \gamma_i \) is the fracture energy of the matrix.

In the MIBF model, crack initiation on carbides is realized as soon as plastic deformation occurs on the matrix. Moreover, no propagation barrier other than the matrix-carbide interface is considered in the model, which means that the fracture of one bainitic packet due to the propagation of a crack initiated on one carbide necessarily implies the fracture of the complete Elementary Volume and eventually of the specimen.

Failure of the specimen from a single carbide of size \( r \) inside \( V_0 \) occurs at a probability \( P_f \) obtained using the critical stress \( \sigma_c(r) = \sigma_c \) in the distribution function \( P(\sigma^* > \sigma_f) \). If we consider a carbide size distribution \( F(r) \), we have to introduce the carbide size density \( \frac{dF}{dr} \) as a weight function, and integrate over \( r \), to obtain the failure probability for one carbide:

\[
P_f(\text{carb}, \Sigma) = \int_0^r \frac{dF(r)}{dr} P(\sigma^* > \sigma_f) dr
\]

Assuming the weakest link hypothesis inside \( V_0 \), failure of volume \( V_0 \) occurs as soon as a crack propagates from one carbide inside \( V_0 \). If \( n_c \) is the carbide density per volume unit, failure of volume \( V_0 \) is given by:

\[
1 - P_f(\text{carb}, \Sigma) = (1 - P_f(\text{carb}, \Sigma))^{n_c V_0}
\]

Applying again the weakest link hypothesis to the whole plastic volume of the specimen finally leads to the global failure probability:

\[
P_f(V_p, P) = 1 - \exp \left( \int_{V_p} \ln(1 - P_f(\text{carb}, \Sigma)) \frac{dV}{V_0} \right) = 1 - \exp \left( \int_{V_p} \ln(1 - P_f(\text{carb}, \Sigma))^{n_c V_0} \frac{dV}{V_0} \right)
\]

For the distribution of the local stress field inside \( V_0 \), three different distributions are considered: a Gumbel and a Weibull distributions as well as a Heaviside function in the case of homogenous stress distribution inside \( V_0 \) (Beremin hypothesis). Gumbel and Weibull distributions are parametrized on FE computations of bainitic aggregates (Libert 2011, Vincent 2010, Vincent 2011). In this paper, the Weibull distribution is used:

\[
P(\sigma^* > \sigma_f) = \exp \left( -\frac{\sigma_f}{k_0 \Sigma_f} m_k \right)
\]

where \( m_k \) and \( k_0 \) are assumed to be constant with loading (Vincent, 2011)

For carbide size distribution, different expressions have been introduced in the model: Jatyatilaka (1977), Lee (2002) and Ortner (2005). In the following application, the three parameter Weibull distribution proposed by Lee is used:

\[
F(r) = 1 - \exp \left( -\frac{2r - \gamma_f}{\beta_r} \right)
\quad \text{and} \quad
\frac{dF}{dr}(r) = \frac{2\alpha_r}{\beta_r} \left( \frac{2r - \gamma_f}{\beta_r} \right)^{\alpha_r-1} \exp \left( -\frac{2r - \gamma_f}{\beta_r} \right)^\alpha
\]

It can easily be seen that parameter \( V_0 \) disappears from eq. (1) in MIBF model. The two remaining parameters are then \( n_c \) which is known from metallographic analyses and fracture surface energy \( \gamma_i \). This last parameter, considered as temperature independent, is the only free parameter of the model.

3. Parametrization

The MIBF model is applied to the “Euro Material A” (22 NiMoCr 3 7) data set. The material mechanical characterisation is presented in Heerens and Hellmann (2002) and was re-analysed recently by Wallin (2012). Its chemical composition is given in Table 1. The Master Curve reference temperature \( T_0 \) is equal to -91°C. Beremin
model has been used to describe this database and has pointed out again the issue of the temperature dependence of \( \sigma_u \) (Andrieu 2012).

<table>
<thead>
<tr>
<th>C</th>
<th>Si</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Mn</th>
<th>Ni</th>
<th>Cu</th>
<th>Mo</th>
<th>Ti</th>
<th>V</th>
<th>Nb</th>
<th>Al</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.22</td>
<td>0.23</td>
<td>0.007</td>
<td>0.004</td>
<td>0.39</td>
<td>0.88</td>
<td>0.84</td>
<td>0.08</td>
<td>0.51</td>
<td>0.001</td>
<td>0.003</td>
<td>&lt;0.005</td>
<td>0.013</td>
</tr>
</tbody>
</table>

### 3.1. Stress-Strain behaviour

At the macroscopic scale i.e. specimen scale for the present application, the temperature dependant stress-strain behaviour of Euro Material A is described by an exponential function:

\[
\sigma = \sigma_Y(T) + \frac{P_1}{P_2} \left[ 1 - \exp \left( -\frac{\varepsilon - \varepsilon_L}{c} \right) \right] \quad \text{with} \quad \sigma_Y(T) = \sigma_u + a \exp \left( -cT \right)
\]

where \( \varepsilon_L \) is the Lüders strain and \( \sigma_Y(T) \) is the temperature-dependent yield stress. The values of the temperature independent parameters are given in Table 2.

### 3.2. Crystal plasticity

Crystal plasticity computations were performed to derive the distribution of principal stress \( \sigma^* \) for an applied loading \( \Sigma \), under different stress triaxialities. Both Gumbel and Weibull expressions have been parametrized using crystal plasticity aggregate computations. Weibull expression was preferred here and the values of \( m_h = 7.0 \) and \( k_h = 1.1 \) give a fair description of the numerical results (Vincent, 2011).

### 3.3. Carbide size distribution

Analyses of carbide size for Euro Material A were obtained from Ortner (2005). The global number of carbides per volume unit is \( n_c = (0.76 \pm 0.24) \times 10^{18} \text{ m}^{-3} \). From the frequency of carbides sizes in each class of radius (Fig. 2a), an experimental carbide size density \( dF/dr \) is computed (Fig. 2b). The distribution of Lee (eq. (2)) is then identified on this experimental curve, at the largest experimental values of \( r \), since the largest carbides are the most nocive for brittle fracture. With parameters \( \alpha_r = 1.6, \beta_h = 0.15 \times 10^{-6} \text{m} \) and \( \gamma_r = 0.01 \times 10^{-6} \text{m} \), Lee distribution is well suited between 0.1 \( \mu \text{m} \) and 0.3 \( \mu \text{m} \) (Fig. 2b).

**Fig. 2.** (a) Representation of the fraction of carbides in each size range; (b) Fitting of Lee distribution on the experimental distribution.
Fracture toughness results for Euro Material A were obtained from the Euro fracture toughness dataset compiled by GKSS and presented by Heerens and Hellmann (2002). Four sizes of Compact Tension (CT) specimen were used for calibration and validation of MIBF model: CT12.5, CT25, CT50 and CT100 specimen. The data base gives toughness values for different testing conditions (specimen sizes and temperatures). Only conditions leading to cleavage without significant ductile tearing are used here; i.e. \( T = -154^\circ C \) to \(-60^\circ C\), for CT12.5 and CT25 specimen and \( T = -154^\circ C \) to \(-40^\circ C\), for CT50 specimen; for CT100 specimen only tests at \( T = -91^\circ C\). Ductile crack extension is less than 0.2 mm when fracture toughness is smaller than about 200 MPa\( \sqrt{m}\).

Simulations of the fracture toughness tests were performed thanks to CAST3M Finite Element code. All CT sizes were meshed in 3 dimensions (Fig. 1). They include 6 layers of meshes in the half-thickness. The crack tip mesh consists of parallelepipses of 50 \( \mu m \times 50 \mu m \) in the cross section. This size is chosen to be close to two times the former austenite grain size of the material. Since crystal plasticity law, carbide size distribution and carbide size density are parametrized, the only remaining parameter to calibrate the MIBF model is the fracture surface energy \( \gamma_f \). Determination of \( \gamma_f \) is done by fitting the \( P_r(K) \) response to the experimental fracture toughness distribution for the CT12.5 at \(-91^\circ C\). A best-fit value is obtained for: \( \gamma_f = 8.18 \) J/m\(^2\). The comparison of MIBF simulations with experimental results at all brittle fracture temperatures and specimen sizes is given in Fig. 3.

MIBF model gives quite good results for the whole brittle fracture domain:

![Comparison between MIBF model results for \( \gamma_f = 8.18 \) J/m\(^2\) and experimental fracture toughness results](image)

Fig. 3. Comparison between MIBF model results for \( \gamma_f = 8.18 \) J/m\(^2\) and experimental fracture toughness results (a): for CT12.5 specimen; (b): for CT25 specimen; (c): for CT50 specimen; (d): for CT100 specimen.
For CT12.5 and CT25 specimen (Fig. 3a & b), we obtain a very good agreement between simulation and experimental results for $-154°C \leq T \leq -60°C$.

For CT50 specimen (Fig. 3c), the agreement extends for $-154°C \leq T \leq -40°C$.

For CT100 specimen (Fig. 3d), the experimental results at $T = -91°C$ are correctly modelled.

Comparisons of numerical iso-probabilities of fracture and the full experiments results are presented in Fig. 4 as a function of temperature. The 50% failure probability (green) curves follow quite correctly the experimental distribution up to 200 MPa√m and the lower part of the transition curve is well predicted by the model.

The curve at $P_r = 1\%$ is a good approximation of the lower bound of the experimental results for the different specimen sizes from $-154°C$ up to $-40°C$, i.e. in a range of 50°C around $T_0$. This result is in agreement with the lower bound determination by Heerens (2002) who have estimated that the lower bound is less than 2.5% of the experimental failure probability.

![Fig. 4. Comparison between MIBF model results for $\gamma_f = 8.18 J/m^2$ and experimental fracture toughness results in $K$ vs. $T$ diagrams (a): for CT12.5 specimen; (b): for CT25 specimen; (c): for CT50 specimen; (d): for CT100 specimen.](image)

5. Determination of the role of the various carbide sizes during fracture toughness loading

One of the possibilities of MIBF is to give an approximate evaluation of the contribution of each carbide size to the failure probability. Indeed, eq. (1) can be reformulated as:

$$\ln \left(1 - P_f \left(\Sigma, V_r \right)\right) = n_c \int_{V_r} \ln \left[1 - \int_{0}^{\infty} \frac{dF(r)}{dr} P \left(\sigma^*(\Sigma, V_0) > \sigma_f(r)\right) dr \right] dV$$
If one makes the reasonable approximation \( \ln(1-x) \approx (-x) \), then:

\[
P_f(V_p, P) \approx n_c \int_0^{+\infty} \frac{dF(r)}{dr} P\left(\sigma(\sum V_0) > \sigma_f(r)\right) dr dV = n_c \int_0^{+\infty} \frac{dF(r)}{dr} \left[ \int_{V_p}^\infty \left[ P\left(\sigma(\sum V_0) > \sigma_f(r)\right) dr \right] \right] dr
\]

Now one can define the contribution to failure of every class of carbide size \( [r, r+dr] \) as the differential quantity \( dG \) where:

\[
P_f(V_p, P) = \int_{r=0}^{+\infty} \left[ \int_{r=0}^{+\infty} \frac{dG(r)}{dr} dr \right] \]

so the population of carbides involved in the failure per unit volume \( [(1/V_p) \cdot (dG/dr)] \) can now be compared to the total population of carbides \( [n_c \cdot (dF/dr)] \). For example for a CT12.5 at \( T = -91°C \) (Fig. 5b), one can find that:

- At the beginning of the test (e.g. at 18MPa\(\sqrt{m}\)) the most numerous carbides involved in the failure probability (i.e. the maximum of the corresponding curve) have a size of \( r \approx 0.6\mu m \). Their contribution to the failure probability reaches \( 10^{-8} \) carbide/mm\(^3\)/µm, to be compared to the density probability of the complete population of carbides \( (n_c \cdot (dF/dr)) \) in Fig. 5 which gives a value of \( 10^{-2} \) carbide/mm\(^3\)/µm for the same size. In other words, only 1 carbide of this size over 10\(^6\) is statistically involved in the failure process.

- When the failure probability reaches 66%, i.e. for \( K_I \approx 125\text{MPa}\sqrt{m} \), failure is mainly due to the carbides of size \( r \approx 0.38\mu m \): even though only 1 over 10\(^6\) of them is involved in failure, their contribution amounts to \( 10^{-3} \) carbide/mm\(^3\)/µm. In comparison, the proportion of carbides of size \( r \approx 0.6\mu m \) involved in failure is now 1‰ but their absolute contribution to failure is only \( 10^{-6} \) carbide/mm\(^3\)/µm.

- For higher loadings, the proportion of carbides involved in failure almost does not change (although the total number increases due to the extension of the plastic volume \( V_p \)).

- The dashed lines on Fig. 5 separate, on the right the carbide sites which can be considered as activated by the classical Beremin mechanism, and on the left, those that are also activated due to the stress amplification generated by the crystal plasticity effect.

![Fig. 5. Comparison between the population of carbides involved in the failure \([(1/V_p)(dG/dr)]\) and the total population of carbides \((n_c \cdot (dF/dr))\), black curve) during loading for a CT12.5 specimen: (a) at \( T = -154°C \); (b) at \( T = -91°C \).](image-url)

On Fig. 5, the curves \([(1/V_p)(dG/dr)] = f(r) \) at -154°C and -91°C are the same for low probabilities (under 1%). When the failure probability increases, the size of the carbides involved in failure becomes, as expected, smaller for low temperature (-154°C) than for the intermediate temperature (-91°C). For higher temperatures, the diagrams remain similar to those at -91°C. The results of the experimental carbide analysis have been reported in Fig.5. At least the upper halves of the \([(1/V_p)(dG/dr)] = f(r)\) curves are above the largest measured carbide size value. In particular, this includes all carbides that fail due to the Beremin mechanism. So to correctly parametrize
the model, in particular to choose the type of law for carbide size distribution \(dF/dr\), it is important to perform the determination of the size distributions on very large number of precipitates; probably on 5 to 10 thousand.

6. Conclusions

A microstructure informed brittle fracture (MIBF) model was successfully applied to the Euro Material data base describing the specimen size effect as well as the temperature dependence up to 200MPa/\(\sqrt{\text{m}}\). The only fit parameter is the fracture surface energy which is temperature independent. Thanks to aggregate computations, the role of incompatibility stresses induced by the crystallography of the bainite has been highlighted. Their effect is to widen the range of carbides implied in the fracture process. However, approximatively one half of these carbides still have a size larger than the largest observed sizes underlining the importance to perform characterization on a very large number of precipitates, typically several thousand rather than several hundred. The precipitates to be considered in these large populations are not only carbides but also carbo-nitrides as well as the inclusions of different types.

The flexibility of MIBF model offers numerous perspectives. Indeed, other mechanisms of failure, such as intergranular failure, competing populations of nucleation sites, extremely rare particles, etc. can be introduced quite easily in the post processor. Some of these applications are under progress, in particular, in the frame of probabilistic safety assessments, the study of fracture at very low failure probability that cannot be reached by experiments.

Acknowledgements

The authors thank the European Commission for partial financial support of this study in the frame of PERFORM60 program (Euratom 7th Framework Program).

References

CAST3M, http://www-cast3m.cea.fr/
 Forget, P., Marini, B., Verdière, N., 2001, Prediction of brittle fracture toughness value of a RPV steel from the analysis of a limited set of Charpy results, Proceedings of 9th International Conference on Nuclear Engineering, ICONE 9, Nice, France
Kantidis, E., Marini, B. Soulat, P., 1992, Intergranular brittle fracture in low alloy steel, Anales de Mecanica de la Fractura 9, 150-153
Vincent, L., Gelebart, L., Dakhlaoui, R., Marini, B., 2011, Stress localization in BCC polycrystals and its implications on the probability of brittle fracture, Materials Science and Engineering A528, 5861–5870