An integrated analysis and interpretation approach for investigating irradiation effects on fusion materials

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Abstract

The determination of the mechanical properties of fusion materials is extremely important to guarantee a safe and reliable operation of fusion components. A number of R&D programs related to the irradiation behavior of fusion materials are worldwide launched to better characterize and select the most adequate materials for the various fusion components. These include the analysis of the mechanical properties and careful microstructural observations with the ultimate goal of a better understanding of the underlying mechanisms. The latter play a key role in improving the capabilities and reliability of radiation damage modeling. To successfully reach such a goal, it is necessary to provide reliable experimental data. However, reliable data do not only mean providing well characterized and documented data using state-of-the-art testing procedures but also other necessary information should be provided to remove ambiguities and minimize the risk of misinterpretation. As illustrated with several examples, it is shown that initially-considered experimental anomalies were removed by using such an integrated approach.

Keywords: ***fusion, tensile, hardness, Charpy impact, ductile–to–brittle transition temperature, fracture toughness, crack resistance, flow localization, irradiation hardening, irradiation embrittlement, microstructure.***

1. Introduction

The design of nuclear components requires important information on the materials resistance. At any time of the whole lifetime of a nuclear component, the intrinsic material properties should not be altered by the thermo-mechanical and environmental conditions such as to jeopardize its safe and reliable operation. A cable that should sustain a weight in a stable position under specific irradiation and temperature conditions should perform this task along its whole lifetime. Not only its strength is important to sustain the weight but also its behavior under time, irradiation and temperature should be known. Therefore, a number of tests should be performed in a testing laboratory on the material in which the cable is made to provide the inherent material properties. It can also happen that the cable was in use for a specific duration under specific conditions and the question raises to know whether this cable can be further used in different conditions (higher weight, higher temperature, …). Basically, for this simple example, tensile and creep properties under irradiation are the key properties to be determined. At the bottom line, the cable can be used for the whole lifetime or should be regularly replaced to guarantee its good operation. This small example illustrates the two typical questions that are usually asked to materials scientists, namely:

1. which material is most suitable for a specific component operating in specific conditions, or
2. what is the component lifetime (or residual lifetime).

The general tendency in the nuclear materials community is to apply a number of well established procedures to evaluate the structural materials performances. To simplify, we can identify basically two important parameters used to evaluate the irradiation-induced degradation of structural materials, the yield strength increase (expressing the material hardening or strengthening) and the ductile–to–brittle transition temperature (DBTT, expressing the material embrittlement). After World War II and since the start of the nuclear electricity power technology, these two properties were the basic ones considered to evaluate the materials degradation. A number of correlations were then established to evaluate these properties from an engineering point of view. In particular, because of the key role played by the reactor pressure vessel (RPV), a large number of these correlations were established on ferritic steels. As a result, hardness–to–yield strength, DBTT–to–yield, DBTT–to–fracture toughness and other property–to–property correlations were established and successfully used. However, as a

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general rule, the effects of irradiation on a large number of materials were investigated in terms of hardening (increase of the yield strength) and embrittlement (increase of the DBTT). In the fusion community, these parameters were also adopted, many fusion materials researchers were originally investigating fission reactor materials. Nevertheless, compared to the RPV embrittlement issue, additional constraints should be considered when considering a fusion environment. Indeed, the fusion materials will experience, in general, much higher neutron doses, higher temperatures, transmutation gases (He, H), … Moreover, because the limited high flux irradiation volumes, usually less than 0.5 liter in the International Fusion Materials Irradiation Facility (IFMIF), only small specimens can be irradiated. Therefore, a number of miniature specimens and testing techniques were developed to determine a number of parameters characterizing the flow and fracture properties of irradiated materials. However, this miniaturization is not straightforward as a number of parameters are affected by the size of the specimen. Specimen sizes are often accommodated to fit the irradiation space requirement. Also, most of these parameters are not physical quantities but rather engineering quantities and their interpretation is not straightforward.

Tests are usually performed in hot cells to characterize irradiated materials. Unfortunately, very often, only limited information is taken from these tests and, important information present in the test record is often disregarded or discarded. The main reason is due partly to the testing standards that are usually applied. Moreover, engineering requirements are limited to basic information related to specific parameters. In other words, the test record path is less important except at specific locations. For example, for the tensile test, the yield strength, the ultimate tensile strength and the elongation are usually reported without specifying the path between these characteristic points. Also, the Charpy impact data are usually reported as the absorbed energy as a function of test temperature without reference to the actual load – time test record, only the integral (energy) is reported.

With the increasingly interest of materials scientists in modeling aspects of radiation hardening and embrittlement, it becomes extremely important to provide reliable data that can be used to evaluate the performances of these models. Therefore, a global or integrated approach should be adopted in order to provide reliable experimental data that can be used for further modeling verifications and benchmarking. However, the two questions raised above still apply and the integrated analysis and interpretation approach also helps the choice of the more appropriate materials.

As illustrated in Figure 1, a specific nuclear component is usually submitted to thermo-mechanical loads, to high temperatures and neutron flux. Ideally, one should test the component in similar conditions as during operation including not only normal operation but also abnormal or accidental conditions. This is of course not reasonable and out of question, not only for economical reasons but also, more importantly, for safety reasons. To respond to the operation requirements, in general, the available material choice reduces very rapidly and only few materials are potential candidates for such application. In order to qualify these materials, in particular in terms of their irradiation resistance, a number of mechanical tests, supported by some microstructural observations are usually carried out in dedicated hot cells to determine their properties. These intrinsic properties are then used in structural integrity calculations and, with the aid of probability assessment methodologies, the lifetime of the component is determined. Of course, it is always the safe side, namely the most conservative, side that is selected.

In this paper, focus is put in the material characterization. More specifically, we concentrate on the potential information that is present in a number of classical tests, like the tensile and Charpy impact test, and how it can be integrated in a physically-based frame to the benefit of an improved understanding and a quality assurance of the experimental data. Among a number of properties, we selected the irradiation–induced hardening and embrittlement as they constitute a key role in structural assessment of nuclear components and a number of illustrative examples to demonstrate the importance of this topic.

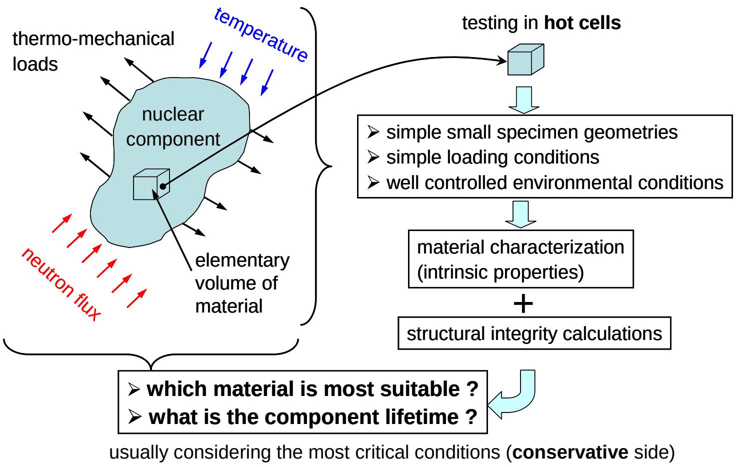


Figure 1. Schematic diagram showing typical questions that are usually submitted to materials scientists in the nuclear field.

2. Irradiation-induced hardening and embrittlement

Basically, when investigating the radiation resistance of structural materials, there are two major words that are used, hardening (or strengthening) and embrittlement. Indeed, as illustrated in Figure 2.a, upon irradiation, the tensile curve exhibit an increase of the stresses and a decrease of ductility. The Charpy impact transition curve exhibits also a shift characterized by an increase of the ductile–to–brittle transition temperature (DBTT) and a reduction of the upper shelf 5Figure 2.b). The correlation between the increase of the yield strength and the increase of the DBTT is illustrated in Figure 2.c which assumes that brittle (instable) fracture occurs when the tensile stress ahead of the notch or crack tip exceeds a critical stress (microcleavage fracture stress) $_c$ over a (metallurgically–significant) critical distance. Brittle fracture is assumed to occur in a transgranular (cleavage) manner. Although not indicated, it is important to mention that if fracture occurs in an intergranular manner, the critical stress may decrease and the DBTT – shift may be larger for a similar hardening. In this case, we refer to non hardening embrittlement. Figure 2 clearly illustrates the inter–relationship between the various properties.

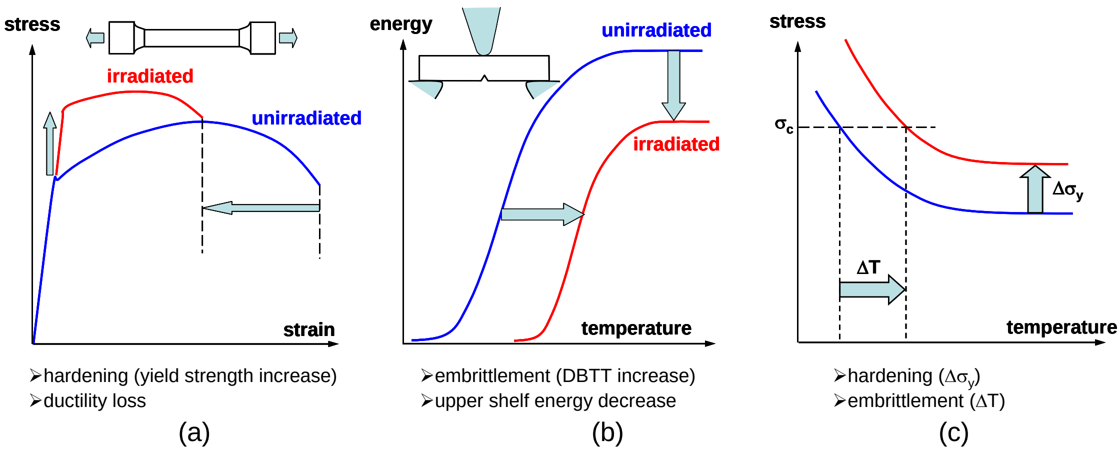


Figure 2. Illustration of irradiation-induced hardening and embrittlement.

2.1. Hardening

From a fundamental point of view, hardening increase is the natural effect of irradiation. Indeed, irradiation induces a number of microscopic features that impede dislocation motion increasing thereby the stress required to overcome these obstacles. This is well described by the so-called dispersed barrier hardening theory. Tensile tests are usually performed on specimens of various sizes and geometries. Normally, there is no size effect on the yield strength. However, very small samples should be considered with particular attention as they can exhibit a much larger scatter. The hardness tests are also often used to determine the irradiation–induced hardening. In Figure 3, a reasonable linear correlation is found between the yield strength ($_y$) and the Vickers hardness measured using a 5 kgf load (H_{v5}). This correlation was obtained on various materials of yield strength ranging between about 100 MPa up to 1200 MPa. Other correlations might be found in literature but these relations were obtained on a much smaller range. The yield strength increase ($_y$) is therefore linearly proportional to the hardness increase (H_{v5}) with a slope equal to 2.89. However, as shown in Figure 3, the scatter is quite large and may bias the actual hardening. Also, hardness tests do not provide full information as the tensile test. For instance, in the two examples illustrated in Figure 4, the occurrence of plastic flow localization (Figure 4.a) or reduction of ductility without change of hardening (Figure 4.b) will not be detected using the hardness test. Therefore, it is important to combine hardness tests with other properties or microstructural information to increase the degree of confidence on these numbers.

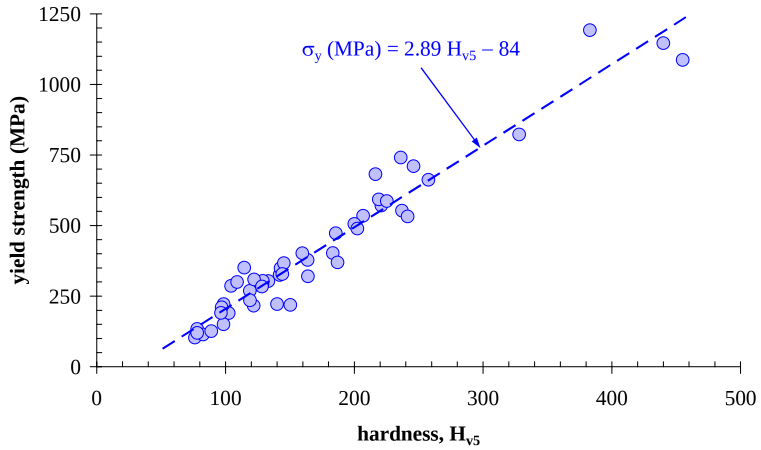


Figure 3. hardness to yield strength correlation.

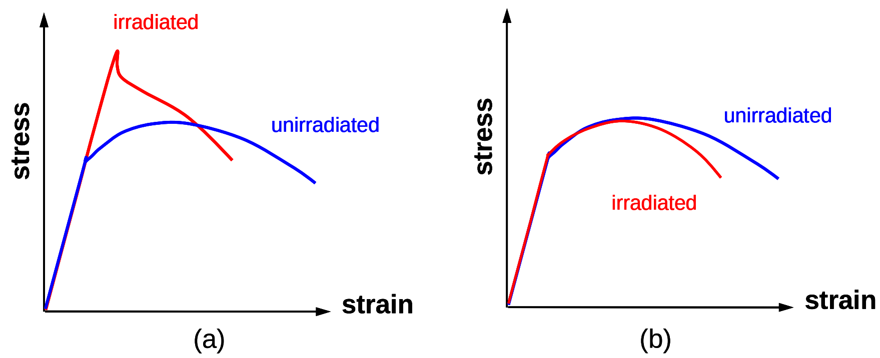


Figure 4. Illustration of the various tensile curves. (a) plastic flow localization. (b) reduction of ductility without hardening.

2.2. Embrittlement

Very often, the irradiation effects are reported in literature mainly as a DBTT–change. However, this is not enough to reliably evaluate the effective irradiation effects. Indeed, is illustrated in Figure 5, a number of situations can be found where interpretation can be biased. For instance, the three cases shown in Figures 5.a, 5.b and 5.c all exhibit the same amount of DBTT–shift. However, there are fundamental differences between these three behaviors that are not captured by the DBTT parameter alone. In Figure 5.a, the DBTT–change is accompanied by an USE–decrease. In Figure 5.b, the DBTT–change is accompanied by a change of transition curve shape in comparison to Figure 5.c. Even the increase of DBTT with a concurrent USE increase can be found, as illustrated in Figure 5.d, in which case careful examination of the data is required. Finally, as illustrated in Figure 5.d, the DBTT–shift can be zero (fortuitously) while the transition curve shape has changed. The list of such “strange” situations is not exhaustive. Nevertheless, it is important when such a behavior is observed to more critically analyze the data and to combine it with other information to reliably assess the material behavior. It is also very important to examine the whole test output (raw data) rather than a global parameter such as the DBTT for example.

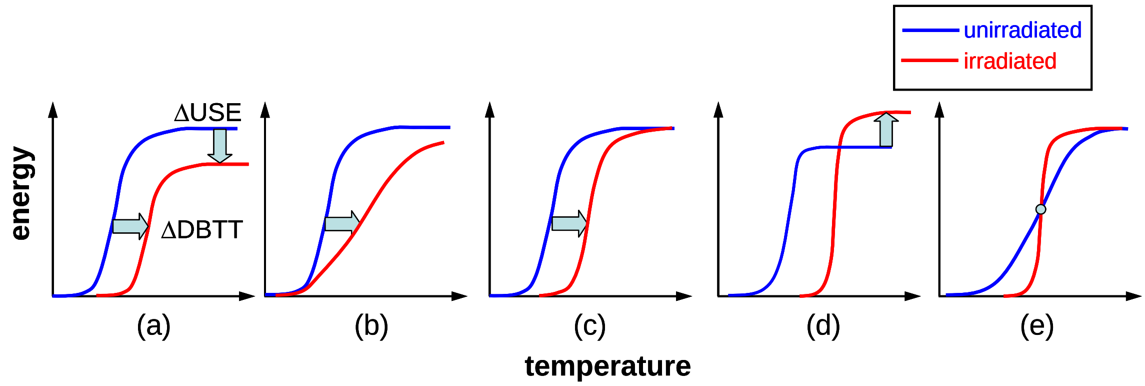


Figure 5. Illustration of the various transition curve behaviors. (a) DBTT–shift and USE energy reduction. (b) DBTT–shift depends on the energy level. (c) DBTT–shift with no change of USE. (d) increase of upper shelf after irradiation. (e) negative or positive DBTT–shift depending on energy level.

As mentioned above, there are basically two parameters that are used to evaluate the degree of embrittlement of irradiated materials, the hardening (yield strength increase) and the embrittlement (DBTT–increase). However, while the yield strength has an intrinsic physical definition and is usually univocally determined, the DBTT is more an engineering quantity of definition that is more questionable. Indeed, the DBTT is usually determined at a fixed arbitrary energy level (Figures 6.a and 6.b). It can also be connected to the upper shelf energy (Figure 6.c). Finally, while reduction of size does not normally affect the yield strength, the DBTT is usually reduced when specimen size id reduced (Figure 6.d). These examples clearly show the limitations related to the use of the DBTT as a parameter characterizing irradiation effects.

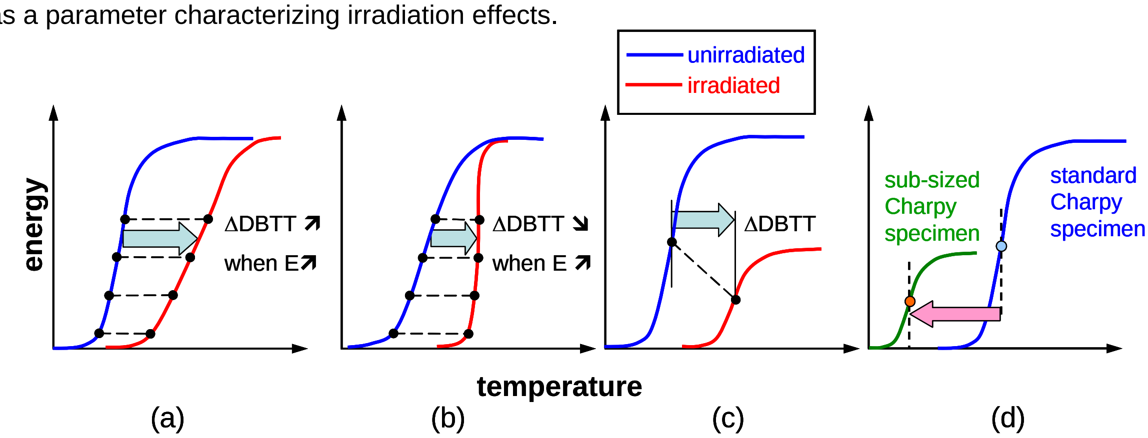


Figure 6. Illustration of the various Charpy transition curve behaviors. (a) increase of DBTT–shift with increasing energy (E). (b) decrease of DBTT–shift with increasing energy. (c) DBTT–shift at 50%–USE. (d) effect of specimen size on the DBTT.

Finally, the DBTT can also be determined using fracture mechanics specimens rather than Charpy V–notch samples. Although the DBTT obtained using Charpy V–notch impact specimens is different from the fracture toughness transition temperature, their respective shifts are usually considered to be equal or alternatively correlated with an additional constant bias. However, there is one important difference between the Charpy V–notch impact transition curve and the fracture toughness transition curve. While the shape of the first one is usually altered by irradiation, the shape of the fracture toughness curve remains usually unaffected. Consequently, the fracture toughness transition temperature shift does not depend on the fracture toughness level at which it is determined. However, as will be seen later, the fracture regime at which the transition temperature is determined should be identical and should not be biased by a change of fracture mechanism with irradiation.

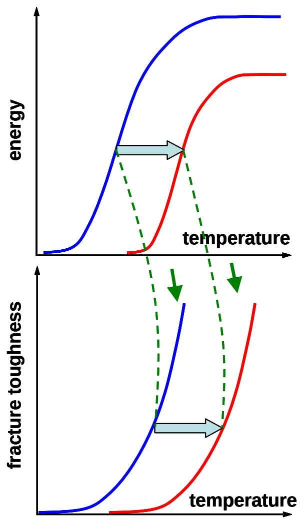


Figure 7. Correlation between the DBTT–change and the shift of the fracture toughness transition curve.

All these illustrations, based on actual observations, indicate the importance of a good analysis and interpretation of the experimental results and the correlations that exist between all these properties.

3. Property–to–property correlations with some illustrative examples

To better illustrate the importance of the property–to–property correlation concept, a number of examples were selected. As it will be seen, examination of one property does not always indicate the actual behavior. This means that, in some cases, examination of a single property can be misleading and incorrect conclusions might be drawn.

It is extremely important to associate mechanical properties to their underlying physical understanding. For instance, the transition temperature is a temperature characterizing the transition from the brittle–to–ductile fracture. However, this is not a physically-based temperature as it depends on the specimen size and configuration. However, even when it is based on a standard specimen configuration, such as the Charpy impact test, the definition of the DBTT can be altered.

3.1. Ductility–to–fracture toughness correlation

The first example consists in examining the term ductility. When investigating irradiation effects of the mechanical properties, a reduction of ductility is usually associated with reduction in fracture resistance. In Figure 8, we selected a 9%Cr-martensitic steel that was tested in a temperature range of [25 – 500 °C] in the ductile regime. In the left side of Figure 8, the engineering strain – engineering stress curves are shown while their crack resistance curves are shown in the right side. As it can be seen, while quite different tensile curves can be observed, the crack resistance behavior does not seem to be influenced by test temperature. This can be understood by examining the fracture surfaces by scanning electron microscopy (SEM) which indicates a different surface morphology. The average dimple size is found to increase with the test temperature. This is shown in Figure 9 for the 25 and 550 °C test temperatures. As a result, ductility alone cannot be unambiguously correlated for ductile crack resistance. Indeed, the material strength also plays a role and the size of the coalescing voids also is important.

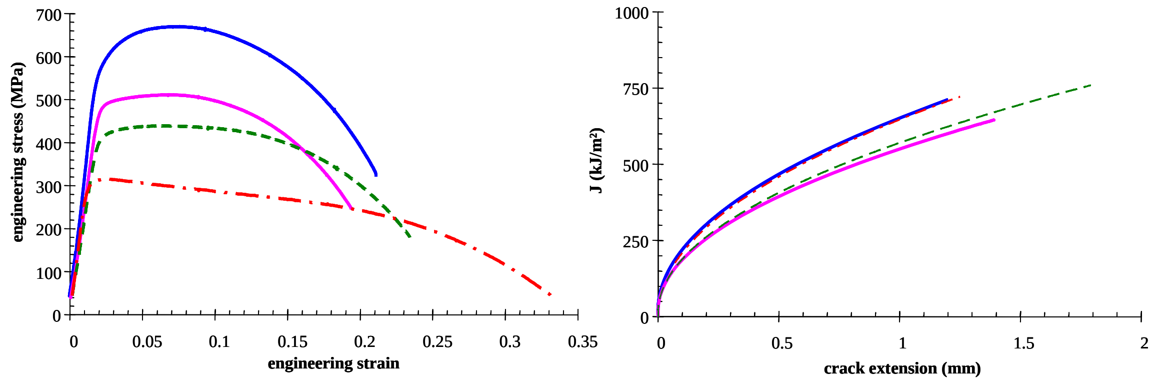


Figure 8. Although tensile properties are exhibiting significant differences, crack resistance behavior remains almost constant.

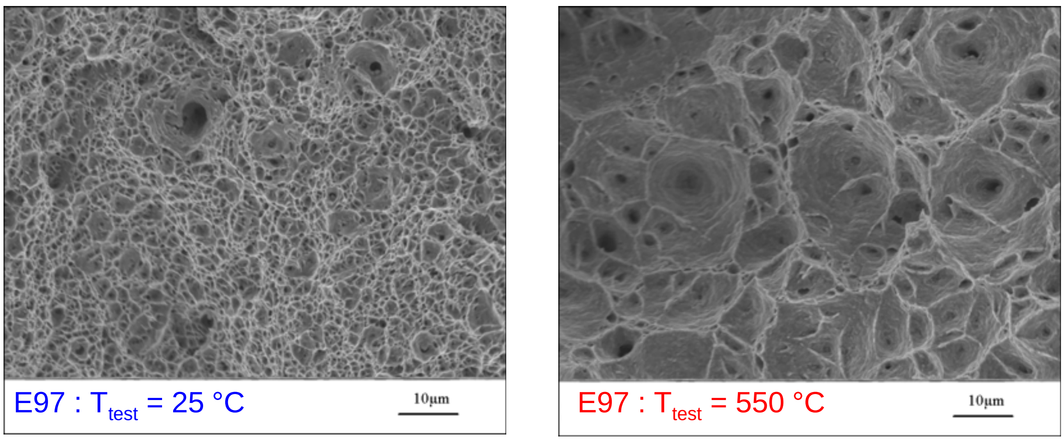


Figure 9. SEM observations indicating increased dimple size with elevating test temperature.

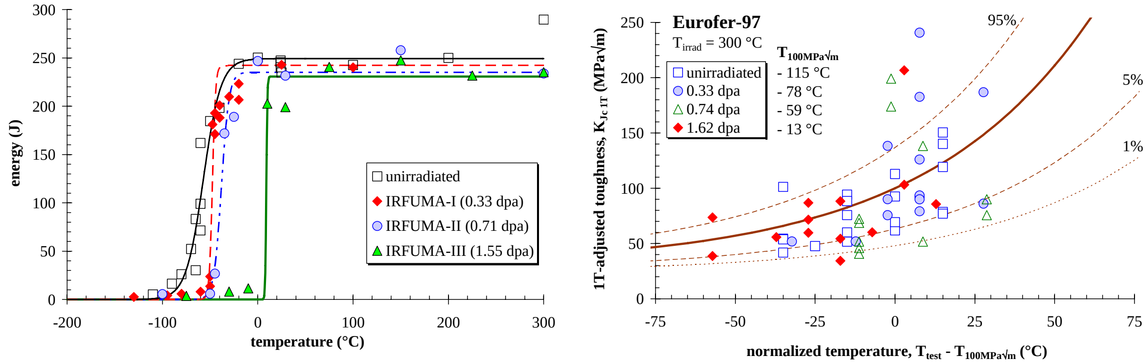
3.2. DBTT–to–fracture toughness transition temperature correlation

With the development of the master curve approach which allows the determination of the fracture toughness transition curve using a relatively low number of small fracture toughness specimens, there is a worldwide tendency to use such a transition temperature to characterize the transition temperature. It is also implicitly assumed that a correlation exists between the DBTT–shift evaluated using Charpy V–notch impact samples and the transition temperature based on fracture toughness specimens, usually determined at an arbitrary level, namely 100 MPa m level ($T_{100MPa\ m}$).

As already mentioned above, and illustrated in Figure 7, the DBTT–shift determined using Charpy V–notch impact tests exhibits usually a good correlation to the shift of the fracture toughness transition curve. However, this is not always the case for two main reasons, namely, the DBTT–definition and the shape of the transition curve. Indeed, there is not a unique procedure to determine the DBTT and several definitions can be found in literature. Moreover, several specimen sizes are usually used which also affects the DBTT. For RPV embrittlement, the DBTT is usually defined at a fixed energy level. For fusion materials, the DBTT is usually defined at the half upper shelf energy. Second, while the shape of the Charpy impact transition curve shape is altered by irradiation, the shape of the fracture toughness transition curve remains unchanged. Therefore, the correlation between the DBTT based on the Charpy impact tests and the one based on fracture toughness measurements cannot be unique. However, materials–specific correlations are available and provide reasonable results.

It is interesting to examine the effect of irradiation on Eurofer-97, a martensitic steel extensively investigated within the European fusion program. This material exhibits a DBTT–shift which is significantly smaller than the shift of the transition temperature, $T_{100MPa\ m}$ (see Figure 10). We could suspect the effect of notch acuity (V–notch versus crack), the configuration (shallow notch versus deep crack) and the loading rate (dynamic versus quasi–

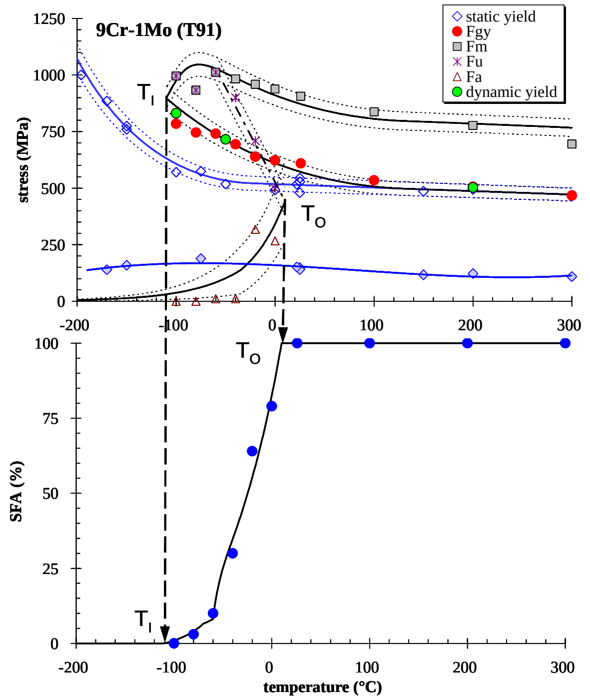
static). However, before questioning these differences, it is interesting to examine the Charpy impact data using the load diagram approach.



(a) (b)

Figure 10. Effects of irradiation on the (a) Charpy impact transition curve and (b) fracture toughness transition curve (master curve).

The load diagram provides a unique tool to describe the flow and fracture behavior of a material. It consists in representing in a constrained physically-based diagram the static tensile together with the Charpy impact data. An example is shown in Figure 11. It should be emphasized that in this diagram, rather than representing the data using an empirical formalism, a physically–based approach is used to reliably associate the actual mechanisms of flow and fracture to the experimental observations. As a result, a number of key parameters can be defined such as T_I, T_O, T_{NDT} and $_c$ (see Figure 11).



T_I : brittle fracture temperature (SFA=0%)

T_O : ductile fracture temperature (SFA=100%)

$T \leq T_I$ → SFA = 0%

$T_I < T < T_O$ → 0% < SFA < 100%

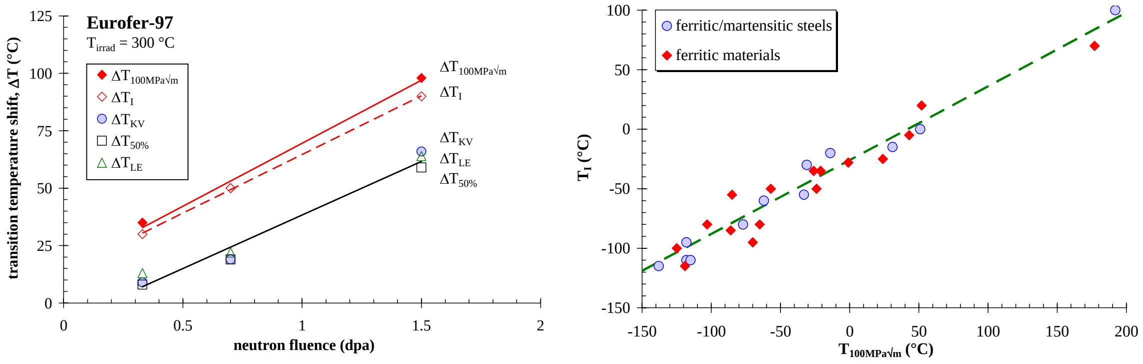
$T \geq T_O$ → SFA = 100%

- $\sigma(T_I)$ → microcleavage fracture stress (σ_c)
- T_{NDT} (based on arrest loads) correlates well to Pellini NDT (Nil Ductility Temperature)

Figure 11. Load diagram of a 9Cr-1Mo (T91) martensitic steel. Illustration of the close relationship between the characteristic loads and the shear fracture appearance (SFA).

At T_I, the fracture is fully brittle, namely, fracture occurs at the general yield. The fracture toughness tests are usually performed in a temperature range where fracture is also fully brittle. A censoring procedure was introduced in the ASTM E1921 standard to treat the samples that exhibit either a high toughness or ductile crack extension before cleavage. So, it can be stated that in both cases, i.e., fracture of the Charpy specimen at T_I and fracture of the fracture toughness specimen around 100 MPa√m are both equivalent. Indeed, Figure 12.a shows that the shift of TI and the shift of $T_{100MPa\sqrt{m}}$) are quite similar, in contradiction with the DBTT–shift. Here, the DBTT was based on the energy, on the SFA and on the lateral expansion. The correlation between T_I (impact) and $T_{100MPa\sqrt{m}}$ (quasi–static) is clearly demonstrated on Figure 12.b for a number of ferritic and ferritic/martensitic steels:

$$T_{100MPa\sqrt{m}}\left(^{\circ}C\right) = 1.79 \times T_I + 70 \tag{1}$$



(a) (b)

Figure 12. Transition temperature correlation between Charpy impact and fracture toughness specimens. (a) Good correlation between $\Delta T_{100MPa\sqrt{m}}$ and ΔT_I in contrast to ΔDBTT (ΔT_{KV}, $\Delta T_{50\%}$, ΔT_{LE}). (b) Good correlation between $T_{100MPa\sqrt{m}}$ and T_I for various materials.

This example clearly shows the importance of associating a physical definition to the DBTT. Indeed, at the usual DBTT, fracture is not fully brittle but it lumps a number of fracture mechanisms including ductile crack initiation, ductile crack propagation, cleavage crack initiation, crack arrest and further shear lip formation. As a result, the comparison between DBTT and $T_{100MPa\sqrt{m}}$ is fundamentally not consistent. Alternatively, the level of impact energy at which the DBTT is determined should be reduced to a level at which fracture is essentially brittle. This way, the brittle contribution is not "polluted" by the other fracture mechanisms.

3.3. USE–to–crack resistance correlation

The Charpy impact test results shown in Figure 10 indicated little to no effect of irradiation on the upper shelf energy. This means that the tearing resistance remains unchanged by irradiation despite clear yield strength increase and transition temperature shifts. When examining the Charpy impact test traces at the upper shelf before and after irradiation, we observe indeed that the total energy is very little affected but a clear difference is obvious before the maximum load is reached (see Figure 13). Indeed, before about 1ms, the load – time traces are significantly different in contrast to the very high similitude of the two curves after 1 ms. The loads are significantly higher than those in the unirradiated condition indicating a significant hardening. On the other hand, static crack resistance measurements indicate a drastic loss of tearing resistance as indicated by Figure 14.a. To remove the possibility of an effect of geometry configuration, namely shallow notch versus deep crack, dynamic crack measurements were performed and confirmed the quasi-invariability of the crack resistance. This means that the loading rate significantly affects the crack resistance behavior. In fact, this "strange" effect can be explained by analyzing all available properties. First, the tensile curves indicate a plastic flow localization occurring above about 0.5 dpa. It is known that this corresponds to another type of deformation called, dislocation channel deformation while slip deformation dominates in the unirradiated (or irradiated to low neutron dose) condition.

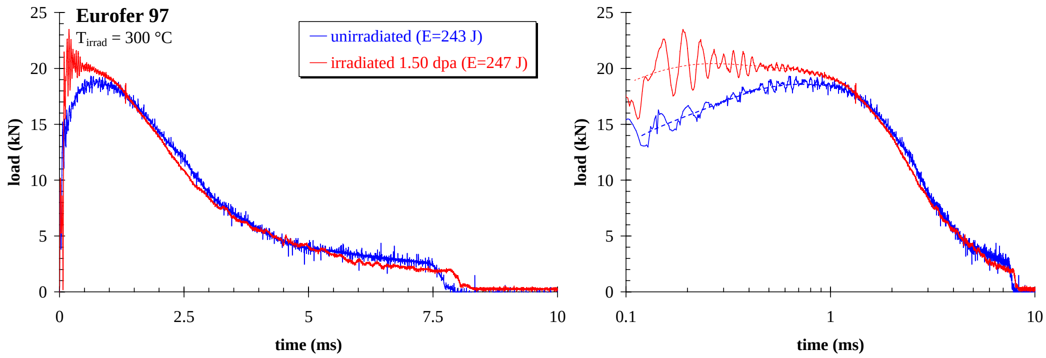


Figure 13. Effect of irradiation on the Charpy impact test record at upper shelf.

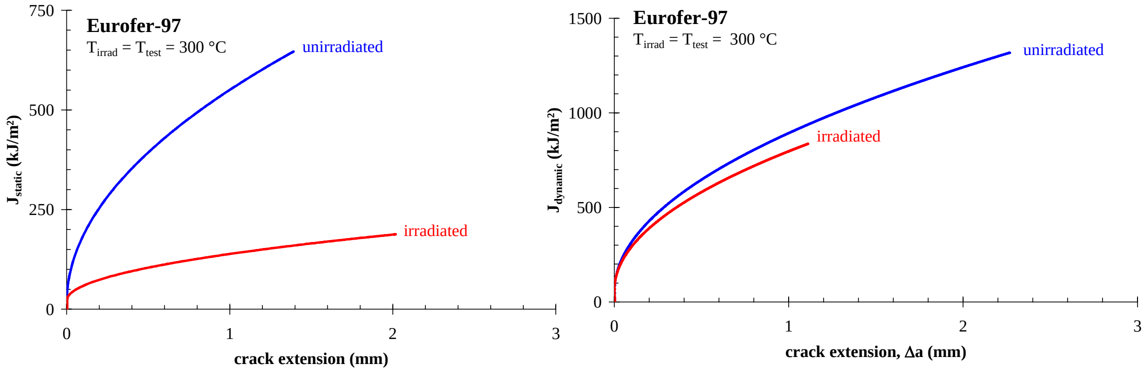


Figure 14. Effect of irradiation on the (a) static and (b) dynamic crack resistance.

4. Discussion

These examples have clearly illustrated the difficulty to consistently and reliably assess the irradiation effects by examining a single property. The available correlations, including both empirical and physically-based correlations, can be helpful to support the final conclusions. However, examination of the raw data rather than the treated data is also of prime importance. Rather than
considering the absorbed energy alone, examination of the load–time test record can bring additional information. There are also a number of consistencies that should be verified when examining the mechanical properties. If hardening is observed on the tensile properties, it should also be observed on the Charpy impact test records. Otherwise, a clarification of this inconsistency is required before data assessment.

Although not directly investigated here, it is very important to mention also an important element related to the specimen size. Indeed, small specimen testing R&D activities were mainly driven by the fusion materials research because of the limitations of the high flux irradiation volumes. There is a natural tendency to miniaturize the specimens as irradiation volumes of intense neutron fluxes are very small, less than 0.5 liter in the case of the IFMIF for example. However, it is important to verify that the mechanisms of deformation and fracture are not modified by the miniaturization. For example, it was shown that in presence of plastic flow localization, static crack resistance is drastically reduced but dynamic crack resistance remains essentially unaffected and one of the reasons was associated to the loss of constraint. Miniature fracture toughness specimens will also experience loss of constraint and that might virtually increase the crack resistance. Therefore, as illustrated in Figure 15, it is important to analyze the mechanical properties and microstructure in an integrated manner to avoid inconsistent conclusions on the actual effects of irradiation. Here, modeling means understanding and it provides a link between experimental observations, both macroscopic (mechanical properties) and microscopic (microstructural observations) and the physics or materials science. The probability of misinterpretation is therefore significantly reduced and experimental biases can easily be detected as well.

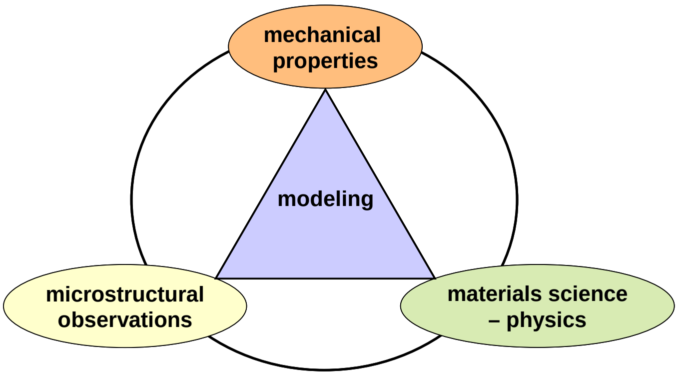


Figure 15. Diagram illustrating the role of modeling in combining mechanical properties, microstructure and physics of materials in a consistent manner.

5. Conclusions

Fabrication of nuclear components, development of new materials aiming to operate in nuclear environments and lifetime assessment of nuclear components all require a number of mechanical properties, in particular under irradiation. A number of these tests are performed in hot cells to investigate the irradiation effects on structural materials. The inherent costs are usually very high, not only their irradiation but also their testing. It is therefore extremely important to extract as much as reliable information from these tests in order to reliably assess their mechanical behavior. A number of parameters are usually used to characterize these materials. For example, the yield strength increase and the DBTT–change are often used to measure the degree of irradiation hardening and embrittlement. However, referring solely to such properties can be misleading. As illustrated by several examples, by combining a number of properties, including microstructural observations and analyzing the raw output data, it is possible to consistently describe the material behavior with a minimum risk of misinterpretation. Reliable physically-based modeling can therefore be obtained in support of the experimental data, providing therefore a kind of quality assurance tool of the experiments.