THE ROLE OF THE BR2 MATERIAL TEST REACTOR TO DETERMINE THE MATERIAL PROPERTIES OF THE FLAKED RPV SHELLS IN SUPPORT OF THE STRUCTURAL INTEGRITY ASSESSMENT

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ABSTRACT

During the in-service inspection in the 2012 summer outage of the Doel-III reactor, a large number of quasi-laminar indications were detected. These ultrasonic observations were, after inspection of the Tihange-II vessel which is very similar to the Doel-III vessel, confirmed a few months later. Those unforeseen in-service inspection results have led to an extensive testing and evaluation program to which the Belgian Nuclear Research Centre (SCK•CEN) made a significant contribution with the determination of the materials properties, required for the safety integrity calculations.
Four irradiation campaigns on a variety of materials relevant for the concerned case were successively carried out under well-controlled conditions in the BR2 Material Test Reactor. Given the high flux of the BR2 reactor, typically more than two orders of magnitude higher than on the vessel wall, the 40 years end-of-life fluence is reached in less than 4 weeks. Thanks to the extensive database developed through few decades of irradiation of a large number of structural materials, the outcome from the BR2 irradiation could be reliably transferred to the reactor pressure vessel.
A number of issues were addressed with in-depth analysis to support the experimental data that were produced. This paper illustrates the key role of the BR2 reactor with several examples taken directly from the experiments performed within this project to demonstrate the applicability of accelerated data to the integrity of actual reactor pressure vessels.

1. Introduction

In summer 2012, an in-service inspection of the Doel-III vessel aiming to detect eventual underclad defects revealed a number of quasi-laminar indications but no underclad defects. Shortly after, given that Tihange-II is the vessel sister of Doel-III, similar in-service inspection on the Tihange-II vessel revealed no underclad defects but, as in Doel-III, a large number of quasi-laminar indications. This confirmed the observations on Doel-III. At that time, the origin and the nature of these indications were not known yet. However, given the location and orientation of the ultrasonic indications, they were very early suspected to be due to hydrogen flaking. This phenomenon is well known in the steelmaking industry since many decades, but it is the first time such a phenomenon is found on a 30 year operating vessel. As a result and according to prevailing regulatory practices, the safety integrity of the vessels should be demonstrated before further operation.

Under the supervision of the Belgian nuclear safety authorities, the utilities launched an extensive testing program to assess the safety integrity of the two vessels. This large program relies on three main pillars:

1. The qualification of the ultrasonic detection technique to ensure that all defects are detected as well as reliably quantified in terms of size, orientation and location [1-2].
2. The material test program investigating the effects of macro-segregation, the effects of ghost lines, the effect of specimen orientation, the effects of hydrogen flakes and the effects of irradiation and thermal ageing [3-4].
3. The structural integrity calculations relying on the two preceding pillars to ensure safe operation of the reactors under normal and accidental conditions [5-6].

It is important to emphasize that both reactors were operating for 30 years without any problem.

In the first step, the main concern was to assess the local properties ahead of the hydrogen flakes. In a second step, an irradiation program was launched to investigate whether the post-irradiation behavior of a flaked material can be affected or not.

As can be expected, a large part of the test program was addressing the post-irradiation behavior of the materials of interest and as such a research reactor capable of investigating these effects in a short time is not only desirable but absolutely necessary. This is how the BR2 reactor of SCK•CEN in Mol, Belgium, played a key role in resolving a number of issues related to the safety integrity assessment of these two hydrogen flaked vessels.

In section 2, the BR2 reactor will be shortly described with particular attention to the irradiation device used in the Doel-III/Tihange-II (D3T2) program. In section 3, we dwell on the reconstitution technology used to increase the database in order to fulfil the large number of regulatory requirements. In section 4, three critical issues are investigated in the D3T2 program: the effect of macro-segregation, the effect of hydrogen flakes involving the measurement of local (ahead of flake tip) fracture properties and, of course, the effects of neutron spectrum and flux which are significantly different in BR2 than at a PWR vessel wall. Finally, the paper will draw some conclusions.

2. Highlights on the irradiation and testing program in the BR2 reactor

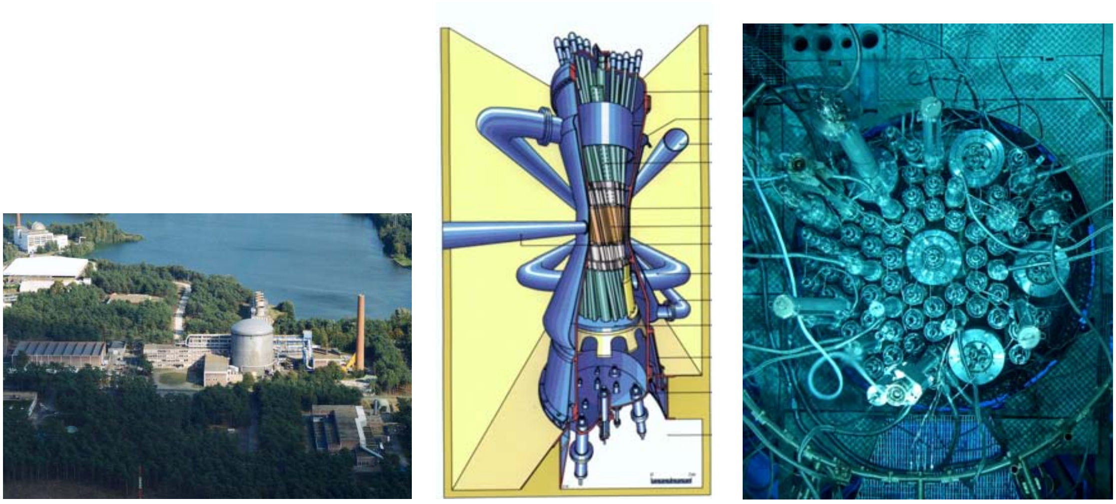
2.1. Short description of the BR2 reactor

The BR2 reactor is a powerful high flux material test reactor. It is operational since January 1963 within the framework of many national/international programs oriented towards structural materials in relevant nuclear environments, nuclear fuel for various types of fission reactors and materials for fusion reactor research. The BR2 reactor also plays an important role for the development and production of isotopes for nuclear medicine, and for electronics and renewable energy applications worldwide.

More specifically, BR2 plays a key role in programs related to:

- the safety of nuclear reactors, plant lifetime evaluation and ageing of components
- the safety of nuclear fuels, the increase of their burn-up and MOX fuels
- the development of new fuels aiming to reduce the risk of proliferation
- medical and industrial applications

A view of BR2 is given in Figure 1a. The key feature of the BR2 reactor (see Figure 1b) is the hyperboloid shape of the berillium-moderated core that drastically increases the neutron density, while the accessibility from reactor top is significantly enhanced (see Figure 1c).



(a) (b) (c)

Fig 1. BR2 reactor: (a) view ; (b) BR2 shape with concentrated core and (c) reactor top view.

In Figure 2, a typical configuration of BR2 at mid plane is shown. The 79 channels allow a variety of tailored configurations to fulfil the customer requirements: the reactor is flexible to accommodate several experimental devices that can be independent from one another. In the case of the BR2 support to the Doel-III/Tihange-II flaked vessel issue, the Callisto pressurized water loop was used (see Figure 3).

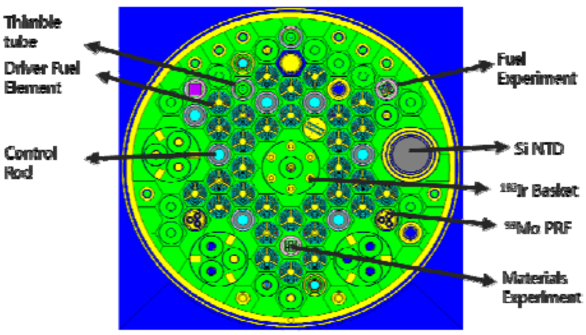


Fig 2. Typical BR2 mid-place cross-section view

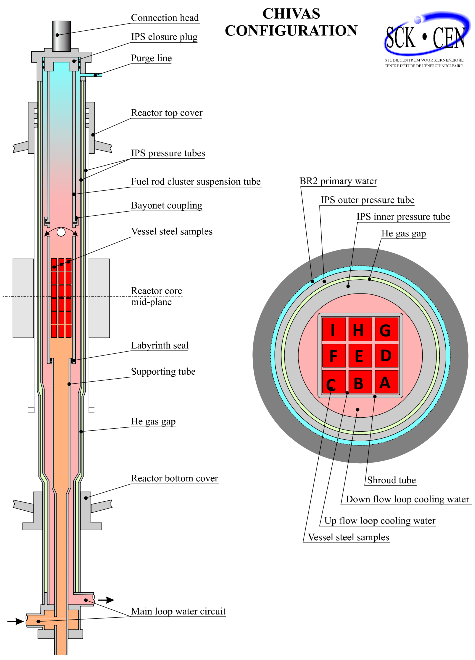


Fig 3. The Callisto loop with a vertical view of an in-pile section withthe basket having nine positions (A - I) for fuel needles or materials

2.2. Short description of the irradiation device

The Callisto in-pile loop with 3 sections was developed in the 90's to replace the fuel experiments after the BR3 reactor shutdown. Since the mid-90's, the in-pile sections were used for both fuel and structural materials irradiation. While the BR2 cooling water is roughly

at 40-50°C, the non-boiling Callisto loop is pressurized to reach a higher water temperature. The loop temperature and chemistry can be easily controlled and the temperature gradient between the center of specimen and its surface does not exceed 3°C. Previous irradiation campaigns were performed at 310°C, 300°C, 285°C, 260°C and 150°C. It is also important to mention that the temperature can be reached before releasing the first neutron on the materials.
The location of in-pile section 3 is at the periphery of the reactor core. This has two consequences: the flux at the mid plane is around 3 10^{13} n/cm²s, E>1MeV and is radially decreasing by about 40%. Therefore, depending on the specifications of the irradiation program, the basket can be rotated 180° at mid-cycle to reach an uniform radial fluence with less than ±3%-deviation. The axial flux shape induces specific fluence levels for the specimens that can then be grouped to obtain specific information at different fluence levels.

Within the framework of the D3T2 test program, four irradiations were performed in the BR2 reactor under the acronym CHIVAS-9 to CHIVAS-12. A typical example is shown in Figure 4 and 5. Here more than one hundred Charpy size specimens distributed over the 9 positions were simultaneously irradiated. The irradiation temperature of the material was 288°C (see Figure 4). The water chemistry was controlled to obtain PWR-conditions: dissolved oxygen, dissolved hydrogen, pH, conductivity,… Given the cosine shape of the axial flux distribution, the specimens experienced different fluence levels (see Figure 5a). A 180°-rotation at mid-cycle allows, when wanted, the lateral fluence gradient to be significantly reduced (see Figure 5b). Dosimetry measurements combined with neutron calculations allow to precisely determine the neutron exposure of each of the irradiated specimens.

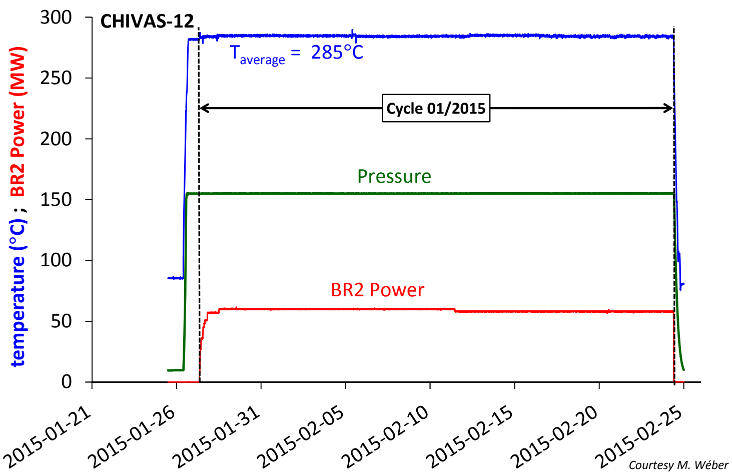
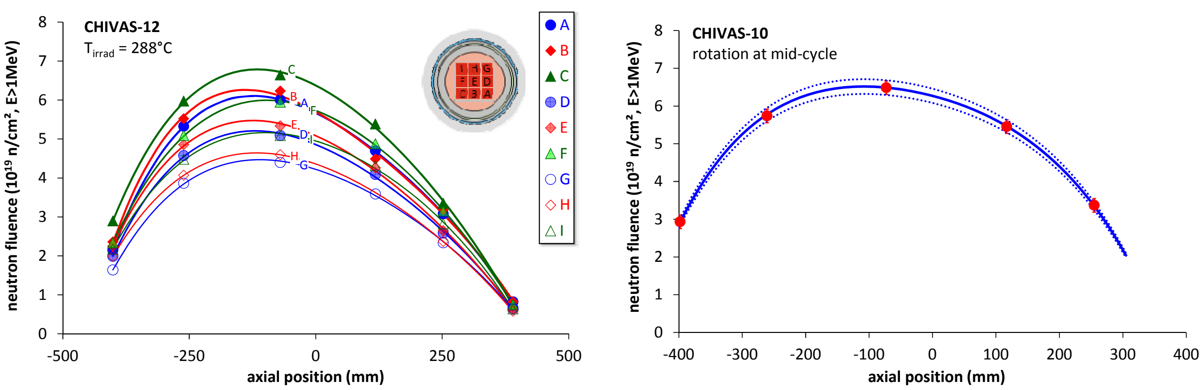


Fig 4. BR2 power and temperature history during the CHIVAS-12 irradiation.



(a) (b)

Fig 5. Neutron fluence distribution of CHIVAS-12 (a) and CHIVAS-10 (b). CHIVAS-10 underwent a 180° rotation at mid cycle for a more uniform fluence distribution (±3%).

The CHIVAS-9 irradiation program specifically addressed a surrogate flaked material, so-called VB-395. More precisely, the fracture toughness properties of the flaked region after irradiation was addressed. The subsequent CHIVAS-10 and CHIVAS-11 incorporated a number of other relevant materials to better calibrate and assess the VB-395 material's behavior. These irradiations confirmed an important non-hardening embrittlement component in the VB-395 material, even in the non-flaked area, a phenomenon that does not occur in the D3T2 materials that were irradiated. Finally, the CHIVAS-12 irradiation program was launched to obtain experimental data on a second flaked surrogate material, the so-called KS-02. Here we wanted to verify whether the non-hardening embrittlement observed on VB-395 is a generic phenomenon that results from the hydrogen flaking process or not. It was concluded that the non-hardening embrittlement is not a generic phenomenon for flaked material.

It is important to state that the well-controlled and known irradiation conditions of BR2 and Callisto allow to assess the mechanical properties of irradiated steels in very reliable manner. A must for using the data in a regulatory based approach.

2.3. Testing optimization

A critical component in the work dedicated to D3T2 was time. Given the economic but also the national importance of the project (Belgium has 55% of its electricity production out of nuclear production and the climate is unpredictable), it was necessary to drastically control the timeframe to accommodate the requirements of all stakeholders. Consequently, a crash program was set up to perform as many tasks as possible in parallel without losing quality control and transparency towards the authorities. The program execution required to work in time shifts because a number of activities can only be executed serially. A typical example of a serial timeframe that was followed, is shown in Figure 6 and this program was executed in a parallel manner for four subsequent irradiations in BR2 with a two to three week gap between irradiations.

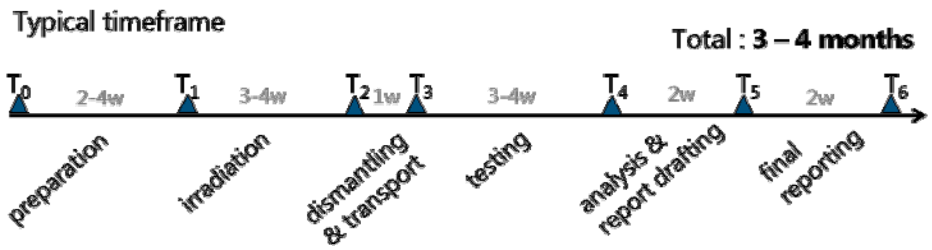


Fig 6. Typical time framework from irradiation to final test results.

Of course, the timeframe also depended heavily on the number of specimens that had to be tested, especially when specimen reconstitution had to be considered to increase statistics. Out of the more than 500 tests performed on irradiated steels, 60% came from reconstituted specimens. As such, we irradiated a total of 228 Charpy specimens from which more than 350 new specimens were reconstituted either for miniature tensile testing, Charpy impact or precracked specimens. In this project, only flat tensile and Charpy size specimens were considered. Figure 7 gives an illustration of the possible specimen configurations from previously broken samples. Reconstitution is available in our hot cells and has been extensively qualified and used.

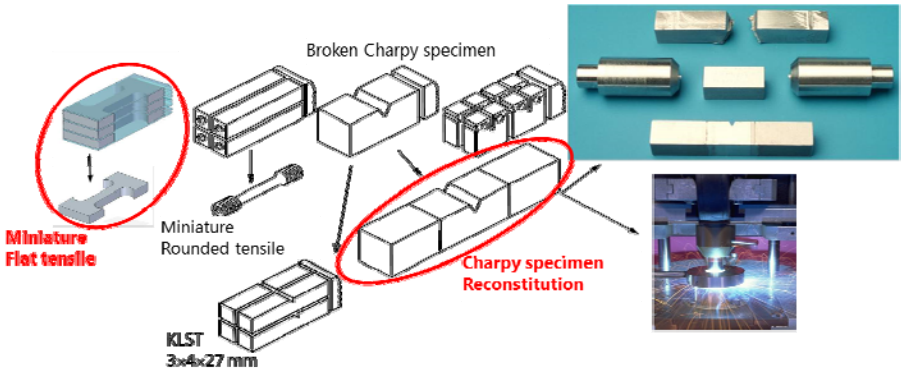


Fig 7. Specimen reconstitution and miniaturisation methods used to increase the number of specimens to be tested.

3. Key experimental data

Within the Doel-III/Tihange-II vessel integrity assessment program, the availability of test data in the irradiated conditions corresponding to 40 year of operation (presently considered end-of-life) were very critical. Indeed, in order to allow operation for the next decennial period, it is required to demonstrate the integrity of the vessels until at least a fluence corresponding to the effective time of operation. Of course, each of the vessels has a surveillance program that monitors the irradiation effects on the material properties with irradiation conditions close to those prevailing at the vessel wall. Typically, the lead factor is about 2.5 to 3. However, the surveillance coupons were taken in an upper or lower shell location that is not affected by hydrogen flaking. Moreover, no significant macro-segregation within the surveillance specimens could be evidenced. Therefore, irradiation of more representative materials that contain macro-segregation and/or hydrogen flakes was required. Moreover, the effect of irradiation on the mechanical properties of those materials upon neutron irradiation levels representative of at least 40-year of operation, typically ~6 10^{19} n/cm², E>1MeV should be investigated. It is obvious that irradiation in a power reactor would require several decades and only a material research reactor is capable of providing such fluence levels in a short time. When using the research reactor, the flux/spectrum differences with a power reactor should be demonstrated to not significantly affect the post-irradiation behavior of the irradiated materials.

As already mentioned, the BR2 reactor was extensively used in previous R&D programs directed towards irradiation effects on RPV materials. This large database was extremely important to provide answers to the flux/spectrum eventual effects. This will be illustrated by few examples taken directly from experiments performed in various reactors. Finally, the paper will address the critical issue on the applicability of accelerated data to the safety integrity of actual reactor pressure vessels.

3.1 Effect of macro-segregation

One of the critical issues addressed in the D3T2 test program is the effect of macro-segregation. The presence of a macro-segregated region is typically found in large forgings as a result of ingot solidification and segregating elements that get trapped in the last cooled region of the ingot (see Figure 8). Typically, the macro-segregated regions are slightly enriched in a variety of elements present in the steel. The macro-segregated regions can be traced by carbon mapping but next to the presence of carbon, all other elements that are segregating, including hydrogen, when present, get trapped in that area. This is the reason why the hydrogen flakes are located within the macro-segregated region. Upon irradiation, some typical segregated elements that are known for high sensitivity to irradiation, such as

Cu, Mn, Ni or P can increase the degree of embrittlement. However, two ways to evaluate the risk on local embrittlement were used in the case of D3T2.

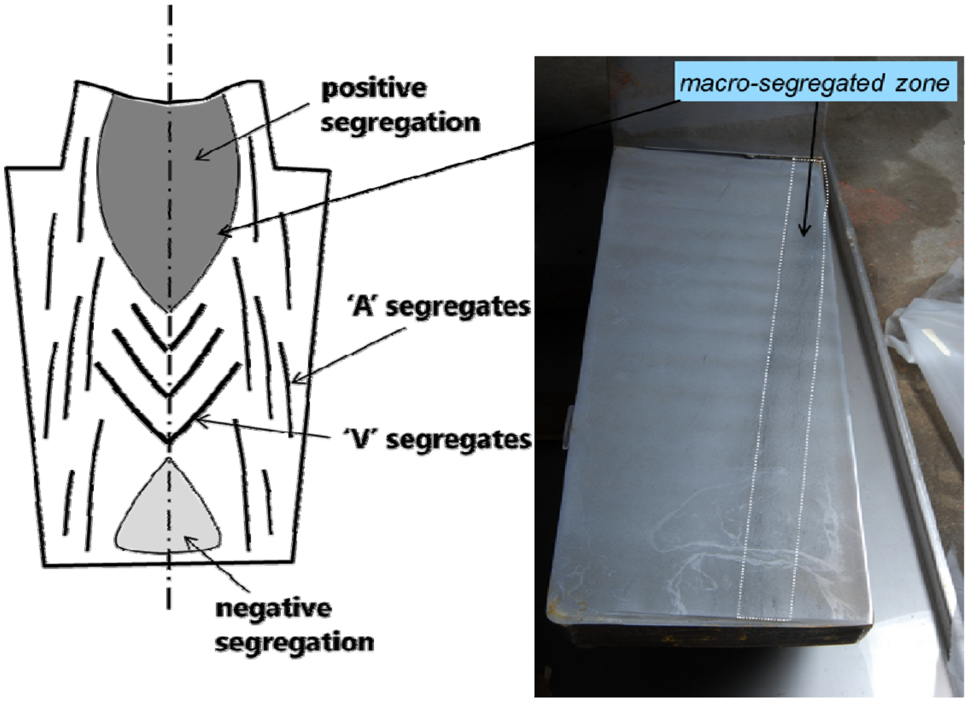


Fig 8. Macro-segregation zone in H1 nozzle cutout forging.

Initially, when no irradiated data on macro-segregated regions of D3T2 were available, but when the concentration of specific macro-segregated elements, known for embrittlement, was measured via microstructural analysis, we could estimate the extra degree of embrittlement by using existing trend curves. These curves, express the embrittlement of a material as a function of element concentration and fluence. By using the measured specific concentrations and plugging them into the trend curves, we can get a conservative estimate of the extra potential embrittlement due to macro-segregation for the D3T2 material.

This methodology was applied in the first safety case submitted to the legislator as no irradiated data on macro-segregated zones were available. As such, the additional embrittlement due to macro-segregation was conservatively evaluated to be less than 17°C.

To confirm this result, specimens carefully taken from the H1 nozzle cut-out of D3, that contained such a macro-segregated region, were irradiated in the BR2 reactor within the CHIVAS-10 program. This irradiation resulted in no measurable extra degradation effect due to macro-segregation in the D3 material. This can be seen in Figure 9 where three sets of Charpy impact specimens in (a) un-irradiated and (b) irradiated to about 6.2 10^{19} n/cm², E>1MeV, are compared. Besides specimens that were respectively taken inside and outside the macro-segregated region of the nozzle cutout H1, a third set corresponding to the surveillance material of D3 and not affected by macro-segregation, is shown. As can be seen on Figure 9, the differences in terms of transition temperature are negligible. These results are confirmed by fracture toughness test results shown in Figure 10. It should be noted that the transition curves based on both Charpy impact and fracture toughness tests for the nozzle cutout H1 outside macro-segregation were higher then inside the zone, but given the limited number of specimens and the small differences, this cannot be considered to be statistically significant.

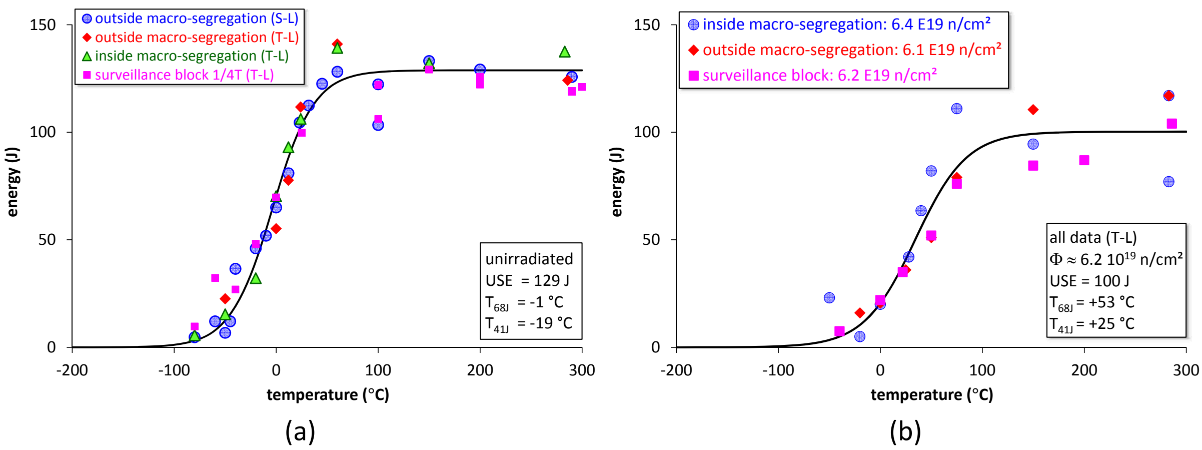


Fig 9. No effect of specimen location (inside and outside macro-segregation) on the Charpy impact transition curve of the materials in both (a) unirradiated and (b) irradiated condition.

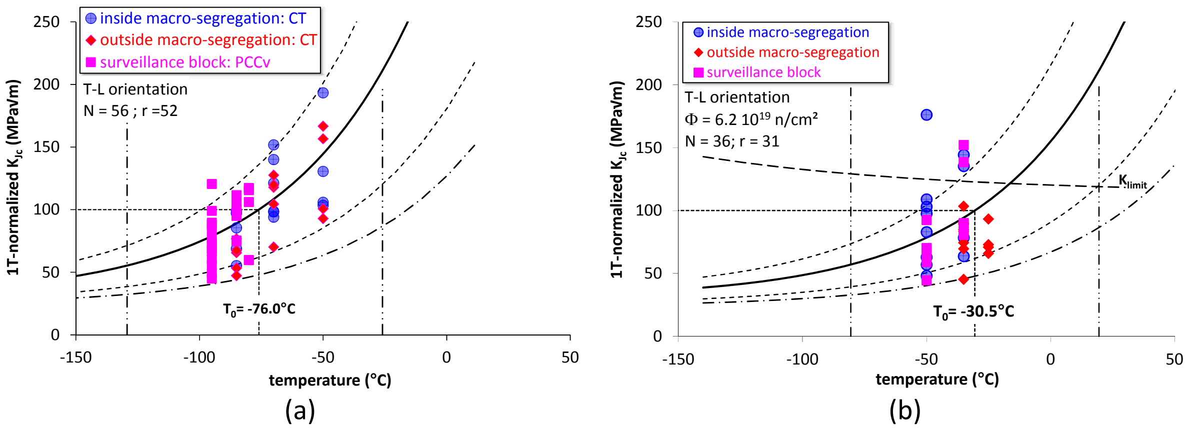


Fig 10. No effect of specimen location (inside and outside macro-segregation) on the fracture toughness master curve of the materials in both (a) unirradiated and (b) irradiated condition.

3.2 Effect of hydrogen flakes

Another key issue that was largely debated between experts is the local fracture toughness properties of flaked material closely near one of the tips of the hydrogen flakes. In order to address this effect, a number of specimens were manufactured in such manner that the hydrogen flake acts as crack starter for fracture toughness testing. In parallel, a series of Charpy size specimen were fatigue precracked according to the E1921 standard and the results are compared in Figure 11.

These two series of specimens were irradiated in the BR2 reactor within the CHIVAS-9 program with a maximum neutron fluence around 7 10^{19} n/cm², E>1MeV. As it can be seen, the two series of data points cannot be distinguished and therefore can be considered as taken from the same population.

More details on this topic can be found in [7].

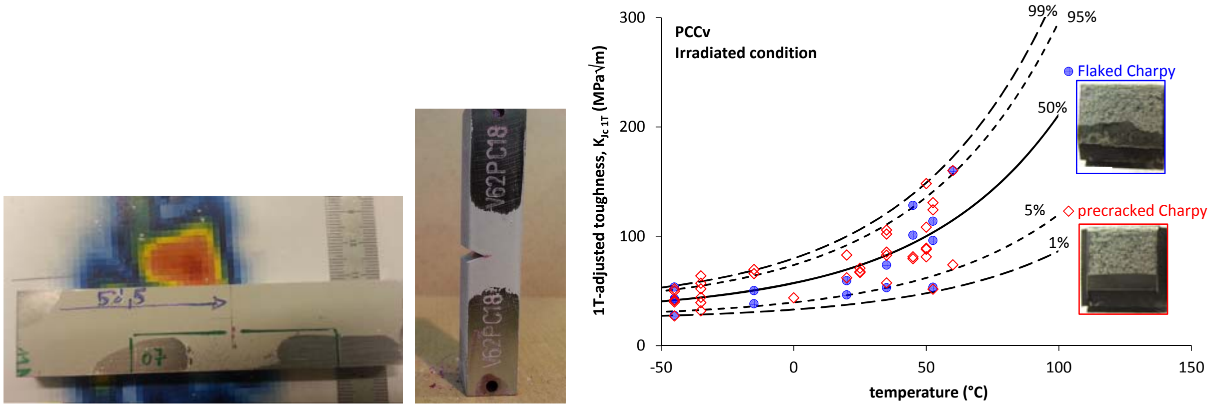


Fig 11. Use of hydrogen flakes as a crack starter replacing the fatigue precrack (a), (b). Very good agreement between fracture toughness data obtained from fatigue precracked and flaked PCCv specimens and associated master curve analysis (c).

3.3 Flux/spectrum effects

The debate on neutron flux/spectrum effects on material properties after irradiation is still not definitely closed and consequently it was also considered in this work. During the last decade, we performed a number of irradiations aiming to assess the magnitude of irradiation hardening and embrittlement resulting from the different flux and/or spectra between research and commercial reactors [8-10]. It should be emphasized here that the separation of flux and spectrum effects is not possible and we consider them to be concomitant. We irradiated several surveillance materials in the BR2 research reactor to compare low flux surveillance data of the NPP's (typically ϕ~1 10^{11} n/cm²s, E>1MeV) to BR2 high flux data (typically ϕ~3 10^{12} to ~3 10^{13} n/cm²s, E>1MeV). An example is shown on Figure 12 for two base metals and two welds. As can be seen, there is no significant effect between the low flux (NPP) and high flux (MTR) data. It is important to notice that small differences are usually observed but they mostly are within the experimental scatter bands.

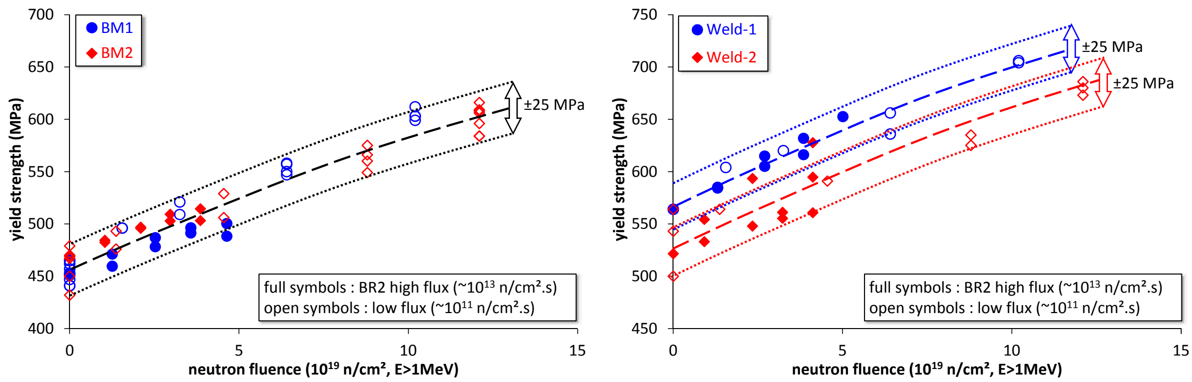


Fig 12. Low flux versus BR2 high flux lead to comparable irradiation hardening.

Another example that was of crucial importance in the D3T2 case, is the irradiation, and subsequent analysis, of the KS-02 material in different reactor systems. The hydrogen flaked KS-02 material was originally irradiated in the FRJ-2 and VAC material test reactors with a neutron flux of about 3 10^{12} n/cm²s, E>1MeV, and subsequently tested at MPA. Recently, in the frame of the D3T2 investigations, we irradiated in CHIVAS-12 three series of 12 Charpy impact specimens of the KS-02 material in the BR2 reactor. These series were each taken from a different location within the unirradiated KS-02 block that we obtained: (i) inside the macro-segregated region between the flakes, (ii) inside the macro-segregated region but outside the flaked area and (iii) outside the macro-segregated region where no flakes are present. Subsequently, the results were compared to MPA series taken from a significantly different location inside and outside the macro-segregated zone. The results in terms of Charpy impact transition temperature shift are shown in Figure 13 [11]. Moreover, we added on Figure 12 two NPP surveillance data points of a 22NiMoCr37 material, that is close in composition and treatment to the KS-02 material, for comparison. As can be seen, all data follow the same embrittlement trend curve indicated by the dotted line.

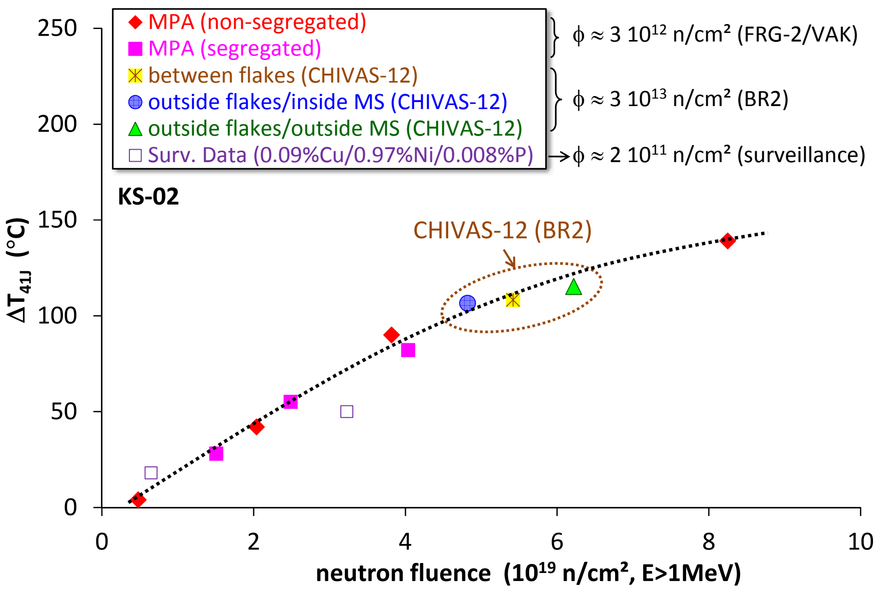


Fig 13. Comparison of irradiation embrittlement data of CHIVAS-12 (ϕ~10^{13} n/cm².s) with those of MPA (ϕ~10^{12} n/cm².s) and surveillance data.

4. Conclusion

In the period 2012–2015, two vessels of the Belgian power reactors Doel-III and Tihange-II were extensively assessed for their structural integrity after the in-service inspection discovery of a large number of indications, that were later identified as hydrogen flakes. Within the extensive test program that followed this discovery, the BR2 material research reactor played a key role thanks to its very high flux but also thanks to the large experimental database accumulated in BR2 material irradiations over the last decades. The BR2 irradiations and subsequent analysis and interpretation delivered the expected results in due time with high reliability.

40-year equivalent NPP neutron fluence on pressure vessel materials can be obtained in the BR2 reactor in 4 weeks. After investigation, neutron flux and spectrum effects were found to be negligible when comparing various data taken from other reactors with inclusion of surveillance data. Within the Doel-III/Tihange-II vessel integrity assessment, it was clearly demonstrated that the presence of macro-segregation does not significantly affect the mechanical properties of the material after irradiation. The local fracture toughness properties ahead of the hydrogen flakes were also demonstrated to be identical to the bulk material properties far from the hydrogen flakes.

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