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Title:	Doel 3 & Tihange 2 RPV issue: FANC synthesis note on "hydrogen induced cracking" hypothesis

Summary:	In complement of the FANC 2015 final evaluation report on the Doel 3 & Tihange 2 RPV issue, the present report describes the HIC hypothesis, its evaluation and the conclusions and the position of the FANC on the impact of this issue.

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Table of contents

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1. Context

1.1. 2012 Licensee Safety Case and evaluation

As part of the 2012 Safety Cases [1] [2], the Licensee made an assessment of the potential growth over time of hydrogen flakes. Based on this thorough analysis, fatigue crack growth had been identified as the only propagation mechanism possible in the specific situation of the Doel 3 and the Tihange 2 RPVs. Other propagation mechanisms (such as defect growth by a mechanism similar to flake formulation due to hydrogen intake from the primary side, and hydrogen induced cracking) were ruled out:

> ***"Defect growth by a mechanism similar to flake formation***
>
> *Creating the same conditions as those occurring during flake formation requires a massive hydrogen feeding. This is impossible:*
>
> *• After the various heat treatments during the manufacturing process and after 30 years of operation at 300 °C, all of the diffusible hydrogen that stems from the fabrication process will have left the material.*
>
> *• The in-service uptake of hydrogen from the primary water or from corrosion processes is very limited (well below 0.1 ppm). This is confirmed by the absence of operating experience feedback on hydrogen-induced cracking in reactors which are or have been in operation. This is especially noticeable for first generation VVER reactors (the Russian version of PWR reactors), which do not have a stainless steel cladding and for which the potential hydrogen intake from the primary environment is higher.*
>
> *• There would be a concentration gradient of hydrogen from the inner to the outer surface, where no hydrogen is present. If this mechanism were active, hydrogen flakes should be larger in size close to the surface than any found deeper in the vessel. This has not proved to be the case. The largest indications are clearly not close to the surface but rather at a depth of 40 to 60 mm.*
>
> ***Hydrogen-induced cracking***
>
> *Hydrogen-induced cracking would require a considerable amount of hydrogen to be present in the steel. This is not the case, as is explained for defect growth by a mechanism similar to flake formation.*
>
> *In addition to a sufficient hydrogen intake in the material, sufficient stresses are also required to propagate the flaws. In operation, the stresses (in the direction perpendicular to the flaw) on nearly laminar flaws are low."*

In May 2013 the FANC concurred with these conclusions in its Final Evaluation Report [3]:

> *"Significant evolution over time of hydrogen flakes due to the operation of the reactor units is unlikely. Indeed, the indications identified are still characteristic of hydrogen flakes even after 30 years of operation. Furthermore, the only theoretical propagation mechanism is low cycle fatigue, which is considered to have a limited effect."*

1.2. Hydrogen Induced Cracking hypothesis

However in the beginning of 2015, some public statements challenged this conclusion. A hypothesis of hydrogen blistering or hydrogen-induced cracking was put forward by prof. W. Bogaerts (KU Leuven) and prof. D.D. Macdonald (UC Berkeley) who state that the exposure of the reactor pressure vessels to the primary water during operation could result in a pressure increase in the detected (hydrogen-induced) flakes and lead to the growth of the detected flakes during the future operation of the RPV [4]. They considered that the issue was not conveniently handled in the 2012-2013 Safety Case evaluation.

The conclusions of a paper written by Bogaerts, MacDonald, Zheng and Jovanovic [5] on this topic are as follows:

"After almost 3 years of investigations, it remains unclear if the cracks found in the Belgian NPPs Doel 3 and Tihange 2 are "only" manufacturing artefacts, or if there is also an "operational component" contributing to the current problems and operational risks; i.e. whether the cracks are still progressing and whether there are other phenomena, e.g. similar to 'hydrogen blistering' processes, contributing to the problem (e.g. some kind of 'delayed cracking' or HIC). Additional hydrogen might indeed come from the cathodic corrosion reactions occurring on the primary water side of the reactor pressure vessel or from other sources such as the radiolysis of the reactor water, or even from nuclear transmutation reactions.

During operation, there is a permanent flux of (corrosion-originating or other) atomic hydrogen through the RPV wall – the flux might be large or small – and this hydrogen could easily get trapped into the voids or "flakes" that are present. An eventual pressure build-up in the flakes will result in growing cracks and other materials degradation phenomena.

Whatever interpretations, disputes, models, theories, etc. there might be, it is a sure fact that the RPV wall will indeed be exposed to significant additional quantities of hydrogen during operation. Significant amounts of hydrogen will also enter (or be generated) within the – already flawed – steel wall.

Such operationally-generated hydrogen will, basically, have two effects:

(1) Either it will cause or aggravate embrittlement of the steel (i.e. also aided by the irradiation);

(2) Or, more direct and abrupt effects may be caused by "loading" the pre-existing flaws, either through the generation of high hydrogen pressures within the voids, or (and) by the building-up of high triaxial stresses at the edges of the flakes, leading to further progressing of the cracks, even with very low external stress levels.

From an engineering point of view this is an unprecedented challenge, especially because of the very high density, the large dimensions, and the great penetration depth of the voids in the metal wall as they have been detected. It is unclear whether this will mean the end of the operation of the reactors, but it certainly is a dominating element in their life management. Other studies need to verify the possible acceptance of the current condition from a mechanical point of view but, if eventually restarted, close-interval surveys and monitoring programs definitely need to be set up for the Doel 3 and Tihange 2 NPPs."

1.3. 2015 Licensee Safety Case

Following these public statements, FANC decided to examine the technical arguments behind this hydrogen-induced cracking hypothesis ("HIC hypothesis") and asked the licensee to deepen its position on this topic.

In response the Licensee performed several additional studies summarized in its 2015 Safety Cases. The hydrogen absorption during operation issue was studied by the licensee, using a numerical model developed by SCK•CEN.

In its 2015 Safety Cases [6] [7], the Licensee concludes that the hydrogen uptake during operation is too low to induce propagation of the existing hydrogen flakes in the D3T2 RPVs by a hydrogen-related mechanism:

"As described in the 2012 Safety Cases and their addenda [1] [2] [8] [9]*, the potential influence of hydrogen uptake from the primary side on the propagation of the hydrogen flakes has been investigated by the Licensee based on literature research, calculations, finite element modelling simulations and hydrogen measurements, as well as consultation with international experts. The assessment has been re-validated in 2015, with an update of the literature review, with newly published results.*

The assessment approach consisted of the following steps:

- *Identification of the possible hydrogen sources from the primary side (corrosion, water treatment, radiolysis).*

- *Quantification of the hydrogen sources (the principal source is the primary H2 water treatment; the total amount is very low).*
- *Calculation of hydrogen accumulation and pressure evolution in the flakes during normal operation and cooldown transients using finite element and analytical modelling.*
- *Evaluation of the risk of propagation due to hydrogen-related mechanisms (hydrogen-induced cracking and blistering) based on the calculated hydrogen content and pressure inthe flakes.*

As the hydrogen content and pressure in the flakes is too low, no primary side hydrogen impact is expected on the material properties and no hydrogen-related propagation mechanism is possible.

The approach and calculation hypotheses have been validated by measurements of residual H content in flakes (showing no significant H content), through literature review and advice from international experts.

The absence of H-induced flake propagation is also confirmed by the absence of evolution of the flakes after one complete cycle and shutdown. "

The Licensee Nuclear Safety Department ("Service Contrôle Physique" or SCP) also made an internal independent evaluation of this topic in its independent reports [10] [11].

"The SCP Review Team has followed the discussion regarding the H2 blistering issue. Several meetings with international experts were held and aimed at exchanging about the possible mechanisms to investigate more deeply. The conclusions of those meetings, also covered by FANC and Bel V, were that the considered mechanisms were sufficiently investigated and that no doubt regarding H2 blistering issue subsists.

The SCP Review Team, in order to challenge the Project Team and experts conclusions, requested the support of Sandia National Laboratory to cross-check the conclusions. After analysis, Sandia National Laboratory concluded that the understanding of the phenomena and the Project's conclusions are correct.

Consequently, the SCP Review Team accepted the Project Team document dealing with H2 blistering."

2. Evaluation by the Belgian Safety Authorities

2.1. Evaluation Process

In this framework FANC and Bel V experts met three times with the Belgian professor supporting this hypothesis. In addition, the Belgian Safety Authorities collected a wide range of material expert opinions on this topic, as described below.

The Scientific Council of Ionizing Radiations treated this subject during their meeting of the 27th of February 2015 and advised the FANC to start a process to investigate this topic.

This process led to the (re)-assembly in March 2015 of the *National Scientific Expert Group* (NSEG) in order to evaluate the relevance of this HIC hypothesis. The members of this working group were selected earlier (in 2012-2013) with regards to their expertise on the subject of material degradation. These members are:

Name	Organism
Rudi Denys	University of Ghent (UGent)
Ludovic Noels	Université de Liège (ULg)
Thomas Pardoen	Université Catholique de Louvain (UCL)
Dirk Vandepitte	Katholieke Universiteit Leuven (KUL)

All these members are independent and have no direct connection with the licensee or the aforementioned professors.

Several expert meetings and hearings with prof. Bogaerts, prof. MacDonald and the Licensee were held by the NSEG, which also received expert reports from three international experts on hydrogen-blistering related issues, two of whom were recommended by prof. Bogaerts himself as renowned experts on the field.

Name	Organism
Peter Scott	Ex – Framatome/Areva
Liane Smith	WG Intetech
Joris Proost	Université Catholique de Louvain (UCL)

To clarify this issue, two workshops were organized by the FANC on 14 April 2015 and on 29 May 2015. The workshops were attended by the members of NSEG, representatives of Electrabel, SCK•CEN, AIB-Vinçotte, Bel V, FANC Scientific Council and the FANC, and invited experts (further termed as Experts) prof. Bogaerts (14 April and 29 May 2015), prof. Macdonald (14 April 2015), prof. J. Proost (29 May 2015), P.M. Scott (29 May 2015), L.M. Smith (29 May 2015).

2.2. NSEG Evaluation and Recommendations

In September 2015, the NSEG issued their Final Report on the hydrogen induced cracking hypothesis [12].

Based on the documents available and the discussions hold during the two NSEG meetings, the NSEG states the following considerations :

> *"Given the operational conditions to be simulated, conclusive experimental investigations to validate the predicted effect of the hydrogen concentration on the pressure increase is technically unfeasible. NSEG also acknowledges that modelling does not provide the desired answer when the input parameters are ill-defined. In particular, a numerical analysis requires assumptions, which might not reflect the actual situation. However, a sensitivity study of the input on the predictions in combination with assumptions made, may lead to a realistic estimate of the actual hydrogen concentration and flake pressure levels.*
>
> *The SCK•CEN numerical analysis, which is based on a rational and well balanced scientific methodology, provides a useful tool for the quantification of the magnitudes of hydrogen concentrations and pressure build-up in the flakes, involving some claimed conservative upper bound estimates. However the NSEG considers that this method has not been fully exploited by the licensee in order to conclude the safety case. More particularly, the assessment of the stress intensity factors resulting from the combined thermomechanical loading and from the hydrogen pressure building up during an unexpected cool-down has yet to be performed and compared to the local fracture toughness for the most critical cracks. Also, the assessment of the conservatism of the approach in terms of the most critical assumptions of the model cannot be made due to insufficiently detailed explanations.*
>
> *Beyond the above considerations, it is worth noting that the size of the flakes has not increased between 2012 and 2014. This demonstrates indirectly that any possible hydrogen uptake had no detectable effect. However, the time of operation between the two successive shutdowns (2012 and 2014) was relatively short and no emergency cool-down occurred during this period. As a result, the number of operational load cycles was too low to provide a definitive conclusion about the absence of crack growth. It should further be mentioned that the crack resistance (of the non-flaked specimens from Doel 3 and Tihange 2, and of the flaked specimens from the other steel blocks which have been measured) may differ from those encountered during operation."*

In consequence the NSEG is of the opinion that:

- *"the licensee has demonstrated that the flakes, which were detected and sized during the RPV UT inspections, using the same and lower threshold settings, are unlikely to have increased in size in the period between June/September 2012 and February 2014. Note that the size of an "individual" flake is smaller than 10 cm as suggested in [3]. The reported defect dimensions were obtained by applying conservative, fracture mechanics based, defect interaction rules ;*
- *at an operating temperature of 300°C, flake growth due to possible hydrogen accumulation inside the flakes can be excluded. At that temperature, (i) the steel of the vessels is ductile (high toughness KIC) and (ii) the H partial pressure is too low (in the order of magnitude of one bar or even less) to have a detrimental effect;*
- *the increase of the partial pressure in the flakes during a cooling transient and the possible H embrittlement (decrease of toughness KIC due to H) requires due attention (see next two points);*
- *the SCK•CEN numerical analysis [8], which is based on a rational and well balanced scientific methodology, provides a useful tool for the quantification of the (a) hydrogen concentration levels and the associated pressure build-up levels in the flakes and (b) the claimed conservative upper bound estimates. However, the stress intensity factor KI values for the maximum partial pressures inside the worst flakes ("worst" in terms of the combination of position in the vessel, size/inclination and local toughness KIC value) in case of a unplanned emergency cooling down following the approach pursued for the safety case analysis , are still to be established ;*
- *the licensee relies on previous tests and on literature to justify that hydrogen embrittlement at the tip of the flakes is not a key issue. However, it be should be demonstrated that (a) the flakes in a fracture mechanics assessment can be treated as "regular" cracks with adequate toughness and (b) that hydrogen damage at the tip of the flake inducing a-typical low toughness KIC will not occur. The study of these specific aspects is beyond the scope of the NSEG analysis, but these issues have been evaluated by the material experts of FANC's International Review Board. "*

Considering the above, NSEG recommends:

1. *"that the licensee should confidently justify that the predicted hydrogen pressures in the existing flakes during cooling are conservative. Further clarification and elaboration of their report [8] are appropriate to provide justification;*
2. *that the licensee should also consolidate its calculation of stress intensity factors in case of an unplanned emergency cooling down by combining the thermo-mechanical loading and the internal pressure evolution. The calculation should be based on the most unfavourable combination(s) of (a) the flake dimensions, their relative positions and flake orientations, (b) local toughness KIC value and (c) conservative (upper bound) hydrogen pressure estimates;*
3. *that regular UT testing is needed to experimentally confirm that the flaws detected in 2012 (and 2014) do not evolve after operations at Doel 3 and Tihange 2 are resumed. "*

2.3. Licensee response to NSEG Recommendations

In October 2015 the Licensee submitted additional reports and calculation notes to the FANC to take into account these recommendations. The Licensee conclusions are [13]:

> *"The hydrogen pressure build-up inside a flake during a 'normal' cooling scenario (from 300°C to 25°C in 24h) was calculated. In addition the impact of an internal pressure of 100bar inside a flake on the crack driving forces of the most critical flakes was calculated. These calculations showed that the impact was small and did not change the conclusions of the structural integrity assessment. This 100bar internal pressure during normal cooling down transient is considered to be a very conservative value, … , the real maximum internal pressure inside the flakes will most probably be closer to or even lower than 1 bar.*

The evolution of the internal hydrogen pressure inside the flakes during a Small Break Loss Of Cooling Accident (SBLOCA) and its impact on the structural integrity assessment were calculated.

Due to the difference in time scales of the two phenomena:

- *Maximum crack driving force due to thermo-mechanical loads occurring at about 1000s after the initiation of the SBLOCA;*
- *Maximum internal hydrogen pressure in a flake occurring after several days;*

it can be concluded that there is no significant impact of the internal hydrogen pressure on the acceptability assessment of the flakes during a SBLOCA.

The FANC and Bel V have evaluated the documents and results provided by the Licensee in order to fulfil the NSEG recommendations. The FANC remarks that the Licensee's additional studies bring the justifications and demonstrations suggested by the NSEG group and strengthen the Licensee arguments considering the non- relevance of this "HIC hypothesis". In consequence, FANC concludes that the Licensee adequately replied to these recommendations.

2.4. Bel V Evaluation

In its 2015 Safety Evaluation Report [14], Bel V concludes that:

"The demonstration of the acceptable serviceability of the RPVs requires that there is no mechanism leading to in-service growth of the flakes. Indeed for the RPV that is assumed not to break, no in-service growth of the pre-existing crack-like defects is allowed. In the 2012 Safety Case, fatigue crack growth was recognized by Electrabel as the only mechanism that could be responsible for potential in-service growth of the flakes (see Section 10). In particular, growth of the flakes by hydrogen-induced-cracking had been rejected by Electrabel as a potential in-service growth mechanism. Occurrence of this mechanism relies on the hypothesis that exposure to PWR primary water can lead to accumulation of molecular hydrogen (H2) in the flakes and to resulting pressure build-up sufficiently high to produce defect growth.

Hydrogen blistering and hydrogen induced cracking are well-known phenomena which can lead to severe damage of a structure or even to its complete failure. It is especially known in the petrochemical industry, which has to carry very harmful fluids with respect to these phenomena.

The phenomena of material damaging relevant to nuclear reactor pressure vessels have been extensively studied since a while. Up to now, these studies did not consider hydrogen blistering or hydrogen induced cracking as an issue for the reactor pressure vessel base metal. This is, amongst others, supported by the favourable return of experience with regard to these damaging mechanisms.

Nevertheless, the risk of hydrogen accumulation in flakes and their consequent growth has been specifically investigated by the licensee in the 2012 Safety Case. The calculations made by the licensee showed that the hydrogen concentrations coming from the different possible sources during operation were too low to cause any hydrogen damage. Bel V agreed with this conclusion.

More recently, following some public statements suggesting still a potential risk of crack propagation due to accumulation of molecular hydrogen inside the flakes, the FANC decided to set up an expert group (National Scientific Expert Group - NSEG) to investigate this specific question. Bel V participated at the meetings of the NSEG.

The arguments presented by the protagonists of the molecular hydrogen accumulation hypothesis did not convince Bel V, neither did they convince the international experts invited by the NSEG. On the contrary, it was concluded that the Doel 3 and Tihange 2 reactor pressure vessels are unlikely to suffer hydrogen induced cracking. As a matter of fact, the hydrogen pressure in cavities within the material cannot exceed the driving force of the dissolved hydrogen partial pressure from PWR primary water under stationary state conditions *at normal operating temperatures, which is insignificant in the context of any hydrogen pressure induced defect growth mechanism. Also, the phenomenon of hydrogen super-saturation due to hydrogen in solution at high temperatures and then quenched rapidly to low temperatures would not be an issue, as the threshold hydrogen concentration for such delayed hydrogen cracking is well in excess of that which could be absorbed from primary water."*

2.5. External experts (Scott, Smith, Proost, Andresen)

In addition to the discussions and debates organized with the NSEG group, the two professors and the Licensee, FANC invited on recommendation of Professor Bogaerts the two corrosion experts Peter Scott [15] and Liane Smith [16], as well as the Belgian professor Joris Proost from UCL [17] to discuss the issue with NSEG and the counterparts and give their independent position on this "HIC hypothesis".

In particular expert Scott makes some specific comments on this issue: although the treatment of the H absorption by the Licensee is incorrect because treating the different hydrogen sources separately is inappropriate, there are significant experiment and theoretical results that demonstrate that the hydrogen activity on the surface cannot significantly exceed that due to the hydrogen fugacity in solution, so that the hydrogen activity cannot exceed $2pH_2$ at 300°C. From Henry's law the fugacity is 2 atm at 25°C and 0.35 atm at 300°C.

For expert Scott neglecting the radiolysis effect as a potentially significant hydrogen source was justified. In addition another potential source of hydrogen within the RPV steel is transmutation reactions yielding hydrogen atoms but this is minor even in austenitic materials relative to the coolant/moderator dissolved hydrogen.

In parallel to the evaluations performed by Bel V and the NSEG group, the FANC explored many tracks and made contact with international experts to identify whether this issue had already been considered in the past. The FANC in particular reviewed on recommendation of EPRI (Electric Power Research Institute) a presentation made by Dr. Andresen from General Electric Global Research Center (GE-GRC) at ICG-EAC [18] in 2015.

In summary, Dr. Andresen concludes that :

1. *Hydrogen flaking in Doel-3 and Tihange-2 RPV raises a number of concerns that require serious thought and investigation.*
2. *H2 permeation in PWR primary water is dominated by the coolant H2 fugacity, and neither corrosion of stainless steel nor radiolysis products has a consequential contribution. (nor does transmutation or proton injection at the RPV).*
3. *H 'pressure' (fugacity) in the RPV and its defects cannot build up over time, and indeed remains below ~0.1 bar (absolute P) because:*
    a. *The coolant fugacity at ~310C has decreased to ~0.3 bar absolute.*
    b. *H2 in defects is not stuck, but can readily dissociate and permeate.*
    c. *H permeation occurs out of the RPV into the surrounding environ.*
    d. *H permeation is greatly limited by the stainless steel cladding. The wall thickness is ~3% of the total, but the permeability in stainless steel is ~200X lower.*

Dr. Andresen's conclusion roughly coincides with Scott's conclusions that the hydrogen pressure in RPV and its defects is not an issue and is limited to some 0.3 bar absolute in operating conditions and 2 bar during cool-down.

3. FANC conclusions

Taking into account

- the initial arguments from the Licensee and the two professors Bogaerts and Macdonald (§1);
- the comments from the NSEG group (§2.2);
- the additional studies and calculations provided by the Licensee to fulfil the NSEG recommendations (§2.3);
- the evaluation by Bel V (§2.4);
- the comments stated by the international experts Scott and Andresen, which are renowned specialists in corrosion effects in nuclear RPVs (§2.5),

the FANC draws the following conclusions on the **HIC hypothesis**:

> *The only theoretical propagation mechanism for the flaw indications in Doel 3 and Tihange 2 RPVs is low cycle fatigue, which is considered to have a limited effect. Other phenomena (such as hydrogen blistering or hydrogen induced cracking) have been evaluated and ruled out as possible mechanisms of in-service crack growth.*

FANC presents this HIC synthesis report and its conclusions to the Scientific Council of Ionizing Radiations for Advice.

In addition, concerning the **evolution of the indications**, the FANC draws the following conclusion based on the Licensee Safety Case and the evaluations by Bel V and AIB-Vinçotte:

> *The evaluation of significant evolution over time of hydrogen flakes due to the operation of the reactor units is unlikely. The comparison between the inspections data from the 2012 and 2014 UT inspections, applying the same parameters and reporting thresholds, do not evidence a crack growth. However, the time elapsed between the restart in 2013 and the shutdown in 2014 is too short to claim that there is a definitive experimental evidence of no in-service fatigue crack growth.*
>
> *Therefore the FANC requires the Licensee to perform follow-up UT-inspections, using the qualified procedure on the entire reactor pressure vessels wall thickness at the end of the next cycle of Doel 3 and Tihange 2, and thereafter at least every three years.*

This additional requirement is consistent with the final conclusions of Prof. Bogaerts and the third recommendation of the NSEG.

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