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Monitoring radiation embrittlement during life extension periods

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HIGHLIGHTS

- Techniques and methods for monitoring radiation embrittlement are described.
- The life extension of the standard surveillance programmes is discussed.
- Guidance is given for the design of new surveillance capsules.
- Recommendations for Integrated and Coordinated Surveillance Programmes are given.

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ABSTRACT

This paper presents guidelines to monitor the radiation embrittlement of reactor pressure vessels (RPV) during life extension periods (to 60 or 80 years) or for the long term operation of nuclear power plants (NPPs). The guidelines were developed in 2012–2013 by a Task Group of the international project LONGLIFE. The work performed responds to the need for guidance to treat long term irradiation effects within the ageing management of NPPs, since the standard RPV surveillance programmes were designed only to cover a time period of 40 years. The guidelines are intended to support specialists in the field and managers in the plant to choose among the most adequate techniques and methods available today to extend the use of their current RPV surveillance programme beyond design life, or implement a new programme when needed. The study performed identifies weaknesses in the ability of the standard surveillance programmes to provide data needed for long term operation, and proposes solutions and tools to solve and/or mitigate the lack or scarcity of surveillance material for their use in life extension. Guidance is also given on methods and strategies to generate reliable surveillance data in the high fluence range.

1. Introduction

Neutron irradiation degrades the mechanical properties of reactor pressure vessel (RPV) steels. The extent of the degradation is governed by a number of factors such as neutron fluence, neutron energy, irradiation temperature, neutron flux and the concentration of deleterious elements in the steel. A RPV operational life of 60 years is being considered frequently by many utilities in their plant life management (PLIM) programmes, and even 80 years is mentioned often in the life extension plans of USA reactors. Guidelines are needed to treat long term irradiation effects within the ageing management plans of nuclear power plants (NPP), in particular for monitoring radiation embrittlement during life extension periods since the standard RPV surveillance programmes were designed only to cover a time period of 40 years. These guidelines should help specialists in the field and managers in plant to choose among the most adequate techniques and methods available today to extend their current RPV surveillance programme, or implement a new

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"Knowledge trasnfer" (Recommendations (3/3) -no.2)

programme when needed, facing long term operation (LTO) of the plant.

This paper summarizes the guidelines for monitoring radiation embrittlement during life extension periods which were developed in 2012–2013 by a Task Group of the international project LONGLIFE. The LONGLIFE project (Project No. 249360 of the Euratom 7th Framework Programme of the European Commission) was designed to enhance knowledge of long term operation phenomena relevant to European Light Water Reactors (LWRs), to assess prediction tools, codes and standards including proposals for improvements, and to elaborate best practice guidelines for RPV irradiation surveillance. The following topics are covered in the guidelines:

- Tools and techniques for surveillance of RPV LTO.
- Extension of the standard surveillance programmes.
- Availability of surveillance material.
- Design of new surveillance capsules.
- Surveillance capsule location.
- Withdrawal schedule of surveillance capsules.
- Thermal ageing.
- Neutron dosimetry and Irradiation temperature monitoring.
- Use of reference materials.
- Use of surveillance data for embrittlement trend curves.
- Use of data from accelerated irradiations.
- Integrated and Coordinated Surveillance Programmes.
- Microstructural analysis techniques.
- Non-destructive techniques.

The guidelines for non-destructive techniques are not discussed in this paper in order to limit the length of this publication.

2. Tools and techniques for surveillance of RPV LTO

This section briefly describes several promising tools and techniques that can be used for embrittlement monitoring during life extension periods. Namely:

"Mini samples"

- reconstitution of broken specimens;
- use of miniature specimens;
- advanced fracture toughness approaches; and
- enhanced surveillance strategy.

2.1. Reconstitution of broken specimens

When plant life extension (PLEX) is required, additional surveillance data are also required, to improve the definition of the embrittlement trend curve (ETC) at the higher neutron fluence levels anticipated. Frequently, however, there is insufficient unirradiated material available to machine additional surveillance specimens for exposure to irradiation for monitoring material degradation during life extension. In addition, conventional (currently-running) surveillance schemes mostly do not include fracture toughness test specimens, even though the determination of a material's fracture toughness is necessary for a structural integrity assessment. Both of these problems may be addressed by the testing of reconstituted broken specimens in suitable specimen configurations. ASTM E1253-07 (2007), Bourdiliaua et al. (2011), Van Walle et al. (2001), and Planman et al. (2012) are important references than can guide in the selection of the most suitable method for specimen reconstitution.

In order to avoid the high costs and burden of long-term storage of irradiated materials, some plants may wish to discard broken Charpy specimens and other surveillance materials remaining from surveillance test programmes conducted years ago. However, broken Charpy specimens, particularly those from reactor vessels having radiation sensitive materials, may provide useful information (such as specific material embrittlement data), and should be stored in appropriate ways and places to avoid oxidation and permit easy retrieval for life extension studies.

2.2. Miniature specimens

Miniature specimen test techniques (Kumar et al, 2006) can significantly enhance the database for assessment of reactor pressure vessel integrity. They provide a means of obtaining material property information for situations in which extraction of samples from vessels (or other structural components) is not desirable or possible, or when the amount of available material is too limited to utilize conventional, standardized techniques. The ASTM books STP-1204 (Corwin et al., 1993), STP-1329 (Corwin et al., 1998) and STP-1502 (Sokolov, 2009) contain an extensive collection of papers on small specimen test techniques applied to nuclear reactor vessel embrittlement, thermal annealing and plant life extension.

Taking into account that the screening limits in the regulations are related to surveillance data from standard sized specimens, it is important to establish correlations between miniature and standard specimen testing results. ASTM E2248 (2009) presents the requirements for performing impact tests on miniaturized Charpy V-notch (MCVN) specimens fabricated from metallic materials. Nevertheless this standard considers that the comparison of the MCVN data with conventional Charpy V-Notch data or application of the MCVN data, or both, to the evaluation of ferritic material behaviour is the responsibility of the user of the test method and is not explicitly covered by ASTM E2248 (2009).

Perosanz (2002) includes an extensive overview of different types of mini-tensile and mini-Charpy specimens used in RPV material testing. Mini-Charpy specimens are grouped in three categories:

- Geometry 1/3: in which the specimen cross section is 3.33 mm × 3.33 mm.
- Geometry 2: in which the specimen cross section is 5 mm × 5 mm.
- Geometry DIN: where the specimen cross section is 3 mm × 4 mm.

Perosanz (2002) and Klausnitzer (1991) reviewed the different methodologies and correlations available to extrapolate results from miniature to standard sized specimens. The correlations should be taken with care since they might depend on the material and the irradiation conditions.

"Master curve"

2.3. Advanced fracture toughness approaches

RPV integrity assessment can be performed using the Master Curve approach (IAEA-TEC-DOC-1631, 2009). In such a case, allowable stress intensity factor values are determined with the use of a reference temperature T_0 (based on static fracture toughness testing of surveillance specimens and/or specimens from template cut from RPV wall) instead of the critical brittle fracture temperature T_k, or adjusted reference temperature ART, derived from T_{41J} (based on Charpy V-notch impact testing). The transition temperature T_0 for the analyzed state of the RPV is determined using a single- or multiple-temperature method in accordance with the ASTM Standard E 1921.

The Master Curve approach is included in different design codes (ASME, KTA) and in the VERLIFE procedure, through the use of a new reference temperature RT_{T0}. ASME Code Case N-631 (Section 3) defines RT_{T0} for unirradiated reactor vessel material, while ASME

Code Case N-629 (Section 11) defines RT_{T0} for unirradiated and irradiated reactor vessel material.

Recently, the fracture toughness temperature dependence $K_{JC}(T)$ for base and weld metals of WWER RPV has been determined according to the procedures given in Russian Standard RD EO 0606-2005 “Procedure for Analysis of In-Service Brittle Fracture Resistance of WWER Reactor Pressure Vessels”. This standard is divided into two parts. Part I is the Basic Curve approach and part II is based on the Prometey Local approach to brittle fracture or Unified Curve. The Unified Curve has been proposed for highly irradiated steels by the Russian CRISM Prometey (Margolin et al., 2005). The special feature distinguishing this model from the Master Curve approach is that it includes a parameter for estimating the effect of irradiation or the degree of embrittlement on the transition curve shape.

2.4. Enhanced surveillance strategy

An enhanced surveillance strategy can be developed for any RPV. A good example comes from the Belgian reactors. For more than a decade, the surveillance programmes of Belgian RPVs have consisted of two complementary approaches. The first one is strictly regulatory following the prevailing standards and codes applicable in Belgium, such as 10CFR §50.61. The second, non-regulatory approach consists of using as much as of the available data as possible in a physically based framework to support the results obtained following the regulatory approach. It often offers additional safety margins with respect to the more conservative regulatory approach. This enhanced surveillance approach is also considered as a quality assurance tool in support of the surveillance programme testing and evaluation. A similar approach is used in the UK.

The Belgian enhanced surveillance approach consists of using instrumented Charpy impact tests to develop the so-called load diagram that incorporates tensile as well as Charpy impact data in a consistent diagram. More physically based transition temperatures can also be determined. Moreover, fracture toughness tests are performed on reconstituted specimens to determine the transition temperature, T_0. Finally, the hardening model allows verification that the experimentally measured surveillance data are well within the predicted trend curves. All this additional information contributes significantly in enhancing the reliability of the RPV integrity assessment, and this is essential in the perspective of long term operation.

3. Extension of the standard surveillance programmes

The standard RPV surveillance programmes are designed to cover the design life which, for American and Western European reactors, is usually a period of 40 years. Nevertheless frequently they have the potential to cover a longer period. Several options should be evaluated by the plant managers to use the available surveillance capsules for life extension:

- Test a different capsule (with a higher lead factor) to that originally planned.
- Irradiate the standby capsules, if available, in positions with a higher lead factor.
- Retain surveillance capsules in vessel for a longer period of time, to increase the neutron fluence accumulated by the capsule.
- Move the surveillance capsule to a higher lead factor position in the vessel. For example, to empty position of previous removed and tested surveillance capsule.
- Reinsert previously removed capsule for additional irradiation (using specimen reconstitution if needed).
- Manufacture a new capsule if archive materials are available.

The withdrawal schedule of the surveillance capsules should be in agreement with the requirements established in Section 7 of this publication. The withdrawal schedule should be reviewed facing changes in the fuel management strategy or due to power uprate taking place during plant life extension. Both plant specific and fleet operating experience should be considered in determining the withdrawal schedule.

The extension of the standard surveillance programme may be insufficient for a proper embrittlement assessment of the RPV. For instance, for 80 years operation the nozzles and the lower head may reach a noticeable neutron fluence (e.g. $>1.0 \times 10^{17}$ n/cm^2 which is the threshold in the German KTA and ASTM E185), and consideration of their embrittlement may become necessary. In this case, the modified surveillance programme should be complemented with the necessary embrittlement monitoring of the “extended beltline material”.

The radiation embrittlement of RPV supports is considered a non-significant issue. The concern is a possible loss of fracture toughness that could result in failure of the RPV supports and consequent movement of the reactor vessel, given the occurrence of a transient stress or shock such as would be experienced in an earthquake. As concluded in NUREG-0933 (2011), prediction of degradation in RPV supports toughness is insufficient to cause concern during 40-year license life. Consideration of a life extension of 20 years did not change this conclusion.

4. Availability of surveillance material

It is frequently found that there is no unirradiated material available for irradiation and testing during life extension. In particular, this situation is very common in the oldest reactors. Suitable alternatives to the lack of archival materials are: (1) the reconstitution of specimens from tested capsules, as explained before and (2) the use of an Integrated Surveillance Programme, as described later in Section 14.

Another possibility, in response to the lack of the original RPV steel, is the use of “tailored” or “surrogate” material. This tailored material is manufactured in such a way that it reproduces as much as possible the chemical composition and fabrication procedure of the original material. This strategy was used by Loviisa NPP Unit 1 in developing the new surveillance programme for operation after annealing (Kohopaa and Ahstrand, 2000).

For 80 years or longer operation times some regions outside the beltline can accumulate a non-negligible neutron fluence, while the concentration of embrittling elements (Cu, Ni, P) in these regions may be higher than in the beltline, or unknown. Materials from regions outside the traditional beltline may not be available for irradiation and testing. Sharing of data between plants of similar design and vendor is vital to address this issue.

The extended beltline regions have a different geometry to the traditional beltline and, therefore, different stress intensity factor correlations. The combination of reference temperature, RT_{NDT} or T_k, and higher stress intensity factor correlations in the extended beltline regions could make these regions more restrictive for pressure–temperature limits after extending operation than consideration of only materials in the traditional beltline. Lack of materials data may lead to a need to assign generic properties for extended beltline materials, and therefore lead to more restrictive pressure–temperature limit curves.

"Expand surveillance program"

5. Design of new surveillance capsules

NPPs may opt to manufacture new surveillance capsules for life extension. The following recommendations are given for the design of new surveillance capsules:

- Make a concept describing the required number of irradiation sets, scope of specimens, withdrawal regime, data assessment, etc., required for RPV irradiation surveillance within a life extension programme.
- Include in the new surveillance capsules materials from the traditional beltline and, if applicable and available, from the extended beltline region.
- As recommended in ASTM E185-10 the surveillance test materials shall include, at minimum, the limiting base metal heat and the limiting weld. For the purpose of determining the limiting materials, predict the adjusted reference transition temperatures for the RPV materials concerned up to the peak fluence of life extension, e.g. according to ASTM E900-02. If a limiting material is outside the beltline, the limiting beltline base and weld materials shall also be included. There is no necessity to include the heat-affected zone material in the surveillance programme as recommended in ASTM E185-10 and KTA 3203–06/01.
- In addition to Charpy and tensile specimens, if enough space is available in the surveillance capsules, include fracture toughness specimens (for instance, for post-irradiation testing according to the Master Curve approach). To increase flexibility for the future testing, the Charpy-sized specimens could be inserted without a notch.
- Include appropriate neutron dosimetry and temperature monitors.
- Ensure that the requirements for the evaluation of the surveillance capsule are met in relation to both national and international regulations respectively, see e.g. ASTM E2215-10.

ASTM E185-10 “Standard Practice for Design of Surveillance Programmes for Light-Water Moderated Nuclear Power Reactor Vessels” does not provide specific procedures for monitoring the radiation induced changes in properties beyond the design life, but the procedure described may provide guidance for developing such a surveillance programme.

Principles for modifying the surveillance programme, or designing a new one if possible, are described in Brumovsky and Kytka (2010a,b) for WWER type reactors. As an example, a combination of archive materials and surrogate materials was proposed for the LTO surveillance programme of Dukovany NPP. Another example is offered by the additional RPV irradiation surveillance programme, with representative specimens of the original RPV base and weld materials, which was implemented in 2007 for the Borssele NPP, in the frame of a LTO programme for additional 20 year life extension (Barthelmes et al., 2010).

6. Surveillance capsule location

Surveillance capsules should be located in positions representative of the irradiation conditions in the RPV wall, in particular taking into account the irradiation temperature, neutron flux and neutron spectrum (Ballesteros et al., 2011). Surveillance capsules should also provide information in advance on the metal condition of the irradiated pressure vessel and, if required, within a certain time period. A good balance between these two tasks is an important requirement for surveillance programmes, and applies to both design life and life extension. The lead factor recommendation generally accepted is between 1.5 and 5. For specific RPV steels used in Germany lead factors up to 12 (KTA 3203-06/01) are accepted. The lead factor is defined as the ratio of the average neutron flux at the location of the specimens in a surveillance capsule to the average neutron flux at the reactor pressure vessel inside surface at the peak fluence location.

Facing life extension, the following strategies can be used to accelerate the irradiation:

- Move the surveillance capsule to higher lead factor position in the vessel, if possible (while remaining within the range 1.5–5).
- Re-design/modify the capsule holders of the operating reactor. This is feasible in some reactor designs (Ballesteros and Jardi, 2008; Caine, 1997).
- Irradiate material from a reactor with poor (low) lead factors in another power reactor with a comparable neutron spectrum and more appropriate lead factors.

7. Withdrawal schedule of surveillance capsules

Section XI.M31 of the GALL report (NUREG-1801, 2010) contains requirements for RPV surveillance programmes during life extension. According to GALL, plant-specific programme should have at least one capsule with a projected neutron fluence equal to or exceeding the 60-year peak RPV wall neutron fluence prior to the end of the period of extended operation. The programme should require the withdrawal of one capsule at an outage in which the capsule receives a neutron fluence of between one and two times the peak RPV wall neutron fluence at the end of the period of extended operation. It is recommended that the programme retain additional capsules within the reactor vessel to support additional testing if, for example, the data from the required surveillance capsule turn out to be invalid or in preparation for operation beyond 60 years.

ASTM E185-10, even if not applying to the LTO of older plants but to the design of new reactors, requires the withdrawal of 5 capsules, after 1/4 EOL, 1/2 EOL (if $RT_{NDT} > 111\,^{\circ}\mathrm{C}$), 3/4 EOL and EOL fluence with testing required, and one standby capsule < 2 EOL fluence without testing (where EOL is the fluence anticipated at end of operational life).

KTA 3203-06/01 follows the pragmatic way of requiring the withdrawal of only 2 capsules, after 50% assessment and 100% assessment fluence, where the assessment fluence is defined as the neutron fluence used in the RPV safety assessment against brittle fracture, and for which the result of the assessment is valid. This is very similar to an approach of withdrawal after 1/2 EOL and EOL fluence, whatever the EOL is (even 80 years and more, depending on the assessment fluence).

However, these basic principles mentioned above may be adjusted to the specific plant conditions since a RPV surveillance programme is mostly plant-specific, including the specific LTO conditions, and has to meet the requirements of national regulations. Such specific LTO conditions are the state of irradiation embrittlement in terms of allowable limits, the availability of surveillance results and number of capsules already tested, the reliability of the extrapolation of reference temperatures for LTO conditions and the EOL time under LTO (usually 60 or 80 years).

It may also be useful to design the withdrawal scheme in such a way that, after withdrawal and testing of any capsule, the next one should be withdrawn before the RPV peak fluence exceeds the fluence of the tested capsule. This is usually the case for PWRs (with higher lead factors than BWRs) and ensures that, after testing the first capsule, surveillance data for experimental verification are always available in advance of operation during the whole operation time.

Based on the international practices the following withdrawal schedule (Table 1) is proposed by the LONGLIFE project:This

Table 1
Recommended withdrawal schedule for life extension.

Sequence	Target fluence	Notes
First	EOL, e.g. 40 years, vessel inside surface	Mandatory capsule
Second	EEOL, e.g. 60 years, vessel inside surface	Mandatory capsule
Third	Between EOL and EEOL, e.g. 50 years, vessel inside surface	Optional capsule
Fourth	1.5 EEOL, vessel inside surface, e.g. 90 years	Optional capsule

EOL, previous end of life; normally 40 calendar years, according to original design.
EEOL, extended end of life.

recommended surveillance programme for life extension is also aligned with the requirements of the GALL report. The first surveillance capsule in the suggested schedule may form part of the standard surveillance programme developed for the design life (40 calendar years) of the plant.

8. Thermal ageing

In most cases it is quite difficult to assess thermal embrittlement based on test results from irradiated specimens. Nevertheless, thermal ageing is not considered in general a problem for RPV steels with relatively low contents of Cu, Ni and P. It is known that Ni enhances thermal embrittlement as a result of activation of segregation mechanisms (Margolin et al., 2012). For WWER-1000 RPV welds with high Ni the embrittlement dependency can be determined on the basis of test results of thermal sets of surveillance specimens, since these specimens are not exposed to any significant radiation. The role of P and Cu on thermal ageing needs to be clarified.

According to the Russian regulatory standard, if the irradiation time is more than 100,000 h thermal embrittlement accounts for a DBTT shift of approximately 20 °C for the Russian steels concerned (Erak et al., 2010). The contributions to embrittlement caused by neutron irradiation and thermal ageing are considered to be additive in this standard.

Nevertheless the time duration under consideration in the perspective of LTO is significantly high, more than 420,000 h for 60 years operation, and it is difficult to unambiguously state whether thermal ageing occurs or not. Investigation of vessel components from decommissioned RPVs, which were outside the neutron flux but subject to thermal ageing, is a valuable method for clarifying the issue. Another alternative is the use of higher temperature to compensate for low ageing time, but this approach should be still validated. It is not recommended that samples be placed in the pressure vessel to monitor thermal ageing during life extension period alone because of the time lag involved.

9. Neutron dosimetry for life extension

Appropriate monitoring of the neutron fluence should be performed for life extension periods. In-vessel or/and ex-vessel dosimetry can be used to this aim. Scraping samples from the RPV wall may be an alternative method for plants needing accurate data concerning the fluence accumulated by the RPV wall, and to support the validation of neutron transport codes.

9.1. In-vessel dosimetry

The standard neutron dosimeters, as used to measure neutron fluence in the original surveillance programme, should be included also in the surveillance capsules planned for life extension. General requirements and possible neutron dosimeters for fast neutron fluences are described in ASTM E-844-09. Nevertheless, experience and lessons learned from the past (Ballesteros et al., 2010) indicate that, in general, neutron fluence values derived from fission dosimeters consisting of ^{238}U, ^{237}Np or ^{232}Th have high uncertainties due to several factors, such as Pu build-in, photofission, etc. Moreover, the fission dosimeters must be shielded from thermal neutrons to reduce fission product production from trace quantities (^{235}U, ^{238}Pu, and ^{239}Pu).

Niobium dosimetry is widely applied instead of fission dosimetry and usually gives reliable results, at least in Western reactors in which high purity Nb wires are used. The ^{93}mNb long half time of 16 years is also favourable in terms of irradiation time. However, high purity Nb dosimeters tend to embrittle after long irradiation/high fluence exposures. Encapsulation may solve the problem because the material is not lost. It remains possible to dissolve the capsule and make the material into a target for X-ray measurements. After decades of experience, the most favoured dosimeter types are apparently niobium and iron dosimeters, but copper and nickel dosimeters may also provide acceptable results.

In addition to the standard radiometric monitors, it is possible to include damage monitors in the surveillance capsules in order to capture both dose and spectral effects simultaneously. Damage monitor sensors are important because they provide a measured response in the neutron energy range between about 0.001 and 0.5 MeV, where radiometric monitors have little or no response. Sapphire, single crystal a-Al2O3 has a number of properties which allow it to be used as a DPA (displacement per atom) monitor.

9.2. Ex-vessel dosimetry

Some plants are opting to use ex-vessel dosimetry to monitor neutron fluence during life extension. Comprehensive sensor sets, mainly radiometric monitors, are installed at discrete locations within the reactor cavity to characterize the neutron energy spectrum variations axially and azimuthally over the beltline region of the reactor vessel, and provide measurements in proximity to critical areas of the RPV. Ex-vessel dosimetry allows long-term monitoring that permits continuous evaluation of the effect of changing fuel management schemes on the RPV exposure.

When used in conjunction with dosimetry from internal surveillance capsules and with the results of neutron transport calculations, the ex-vessel neutron measurements allow the projection of embrittlement gradients through the reactor vessel wall with a low uncertainty.

Ex-vessel dosimetry is unlikely to be used in WWER-1000s for adjustments together with the results from in-vessel dosimetry because of the bad location of the in-vessel surveillance capsules above the core. Methods need still to be developed to correlate in-vessel and ex-vessel dosimetry data for this type of reactor (Ballesteros et al., 2010).

In Western European reactors most of the installed ex-vessel dosimeters, if any, are already tested. Ongoing life time extension programmes of some older plants e.g. (Barthelmes et al., 2010) are managed without any external dosimeters. This is the case in Switzerland, Netherlands and Germany. In contrast, Spain and Sweden currently use ex-vessel dosimetry to complement the standard surveillance programme.

If ex-vessel dosimetry is applied, LONGLIFE recommends a maximum frequency of 5 EFPYs for removing, analysing and replacing ex-vessel dosimeter sets. Shorter periods may be adequate depending on the particularities of the plant. The frequency may be changed if there are significant changes in plant operation. For instance, changes in fuel management strategies (low leakage core, use of MOX, etc.) and power upratings. The frequency of analysis is also set by the nature of the dosimeter materials incorporated in the ex-vessel surveillance programme.

The Ringhals AB approach for ex-vessel dosimetry is described as an example. According to Ringhals AB experience, to obtain the

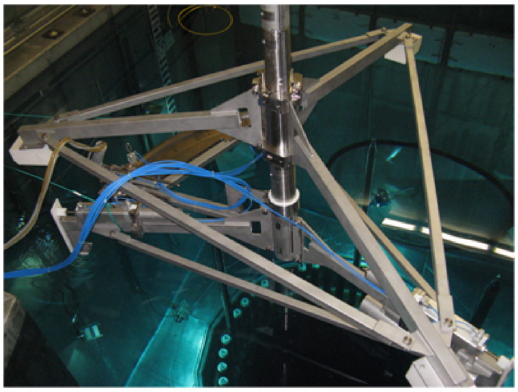

Fig. 1. Carrier for the milling unit for removing scraping samples out of the RPV cladding.

best results from activity measurements from the dosimeters, a set should be exposed for 3–4 years of operation. Once the calculational model for the RPV has been verified by the dosimeter sets, it is not necessary to keep dosimeter sets installed at all times. Ringhals AB strategy for their PWRs is to make a base-line measurement of one set of dosimeters and then make follow-up measurements with a set of dosimeters (exposed during one cycle) in due time before performing a Periodic Safety Report, or when there will be significant changes in core loading or operation conditions. Different but equally good approaches are followed in other European countries (e.g., in Spain and Czech Republic).

For reasons of the high validation level of state-of-the-art neutron transport codes like DORT 4.2, TORT and MCNPX used for neutron fluence calculations, the use of external dosimeters may be waived if results from internal dosimeters or scraping samples are available.

9.3. *Scraping samples from the RPV cladding*

A further method to monitor the neutron fluence is the analysis of scraping samples removed from the RPV cladding. The theoretical calculated neutron fluence values of the RPV, as well as those of the previous irradiation sets, could be verified by means of evaluation of the Fe and Nb inside the cladding activated by neutrons. Particularly in the context of long term operation this method provides reliable results, since there is no embrittlement of pure Nb at long irradiation times. Scraping sample dosimetry may be used to complement in- or ex-vessel dosimetry, and is an appropriate alternative if no reliable results from in- or ex-vessel dosimeters are available.

A milling unit is used to remove scraping samples. This unit may be fixed on a centred manipulator mast, if available. Alternatively a special carrier may be used to fix the milling unit, e.g. as shown in Fig. 1 used in a Siemens/KWU plant.

The withdrawal positions for scraping samples should be defined in such a way to cover the region of the RPV with the highest neutron irradiation (core midplane) and to verify experimentally the azimuthal fluence development along a 45° sector, the 90° symmetry and the axial distribution of the neutron fluence.

Retrospective dosimetry methods were developed in the international project RETROSPEC (Voorbraak et al., 2003). The project focussed on niobium dosimetry. The process involved the extraction and counting of niobium present in the reactor material (cladding and base material). The problem was tackled by taking some scrapings or chips from the reactor material and chemically separating the niobium to allow its activity to be measured. Various chemical separation procedures were explored, and after separation, inductively coupled plasma-mass spectroscopy and neutron activation analysis were considered as proper means to measure the niobium concentration. Counting of aqueous niobium samples, low-mass niobium deposits and liquid scintillation counting were also applied in the RETROSPEC project to measure the niobium activity.

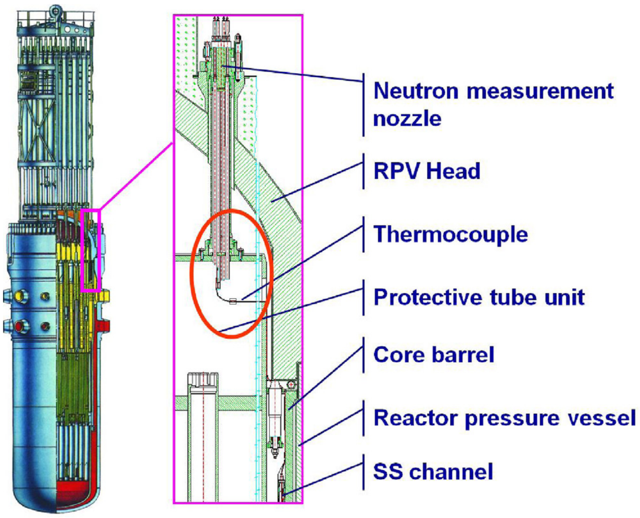

Fig. 2. Irradiation temperature measurements by thermocouples in the COBRA project.

10. Irradiation temperature monitoring

Temperature monitors, usually made of low melting eutectic alloys, are used in LWR surveillance capsules to verify the estimated values of the maximum irradiation temperature of the surveillance specimens. Temperature monitors are needed to give evidence of overheating of surveillance specimens beyond the expected temperature. Since overheating causes a reduction in the amount of radiation damage to RPV steels by annealing, this overheating could result in a change in the measured properties of the surveillance specimens, which would lead to a non-conservative prediction of damage to the reactor vessel material.

The melting monitors commonly used in the standard surveillance programmes can also be used for life extension purposes in new surveillance capsules. ASTM E1214-11 provides guidelines for the selection and calibration of temperature monitor materials; design, fabrication, and assembly of monitor and container; post-irradiation examinations; interpretation of the results; and estimation of uncertainties.

Direct measurement of the irradiation temperature by thermocouples during reactor operation, as shown in Fig. 2, is the most reliable way to obtain this information (Ballesteros et al., 2005). This is a complex engineering task and installation of thermocouples is not always possible. When melting monitors are providing contradictory information, the possible use of direct measurements by thermocouples should be considered. Due to the complexity of equipment installation and associated costs, it should be sufficient to perform direct measurements a single cycle.

11. Use of reference materials

The use of a reference steel, also frequently called a correlation monitor material, can provide an independent check for deviations from the expected surveillance capsule irradiation conditions (for example, temperature, neutron flux and neutron energy spectrum). The use of reference materials like the IAEA JRQ (IAEA-TEC-DOC-1230, 2001) or other well-known steels (Stallmann, 1988) for inclusion in the surveillance capsules is a valuable option, however it is no longer explicitly required in ASTM E185-10.

The JRQ material has been irradiated in some cases to very high neutron fluences and its embrittlement trend curve is very well defined. This steel can act as a correlation monitor material for quality assurance purposes. The surveillance data for the correlation monitor material in the capsule, if present, must fall within the scatter band of its database.

12. Use of surveillance data for embrittlement trend curves

The embrittlement assessment of a specific RPV can be based on two different strategies:

- Using the surveillance data of the specific reactor directly or
- Using semi-empirical embrittlement trend curves generated from large sets of data coming from different reactors. The specific surveillance data is used only to verify that they follow the general tendency.

Both strategies are considered adequate. It is expected, in principle, not to have many surveillance data covering life extension periods before the life time extension programme starts. The statistical significance of the surveillance data from a single, specific reactor would be therefore low. It would be better to check whether the surveillance data of the specific reactor follow the general tendency of a qualified generic embrittlement trend curve with sufficient margins, and to use this curve for the embrittlement assessment of the RPV. It is important also to verify the credibility of the specific surveillance data. For example, credibility criteria for surveillance data are given in Section B of the USNRC Regulatory Guide 1.99 Rev. 2 (RG 1.99-Rev2, 1988) which may be used for the determination of ΔT41 based adjusted reference temperatures (ART) and upper shelf energy (USE).

Generic embrittlement trend curves covering life extension periods can be generated using Coordinated Surveillance Programmes, as described in Section 15 of this publication.

There now exists an extensive amount of new BWR surveillance data from a BWR supplemental surveillance programme (GE, 1989; GE, 1992) designed to augment the plant-specific BWR surveillance programmes. These recent data allow a statistical check of the existing embrittlement correlations.

13. Use of data from accelerated irradiations

Materials test reactor (MTR) data are essential to show the individual effects of radiation and material variables on degradation at high fluence, in order to interpret trends observed in surveillance datasets. In particular, they are expected to give warning of any catastrophic ageing phenomena above a fluence threshold (even if a potential flux effect issue remains). MTR data are thus necessary as a first step towards lifetime prolongations unless appropriate surveillance results are available (Todeschini et al., 2011).

The direct use of test data from materials irradiated in research reactors for embrittlement assessment in power reactors is considered still an open issue (Williams, 2011). For example, a comparison of embrittlement trend curves for low Cu SA-533 Grade B Class 1 Plate shows that using test reactor data at high fluence levels (high flux and high fluence) may lead to non-conservative prediction of the metal condition (Hardin, 2011a). Conversely, according to Soneda et al. (2010), high Cu material irradiated in a MTR appears to show larger shifts than those of surveillance data, while low Cu materials show similar embrittlement. However, the irradiation experiments with German RPV steels in the VAK test reactor did not reveal any significant flux effect even for high copper welds in the mechanical properties (Hein and May, 2008) even though a flux effect was clearly visible in the microstructure (Bergner, 2008). The precise MTR irradiation conditions under which MTR data are conservative/non-conservative for RPV conditions remain to be determined.

Important parameters for the extrapolation of results from accelerated irradiations to typical power irradiation conditions are the irradiation temperature, the neutron flux and the neutron spectrum. In particular, the effect of neutron flux on embrittlement behaviour is considered a complex phenomenon, which itself appears to be dependent on the steel composition, the neutron flux and fluence range and the irradiation temperature.

Nanstad et al. (2008), reviewing a draft NUREG Report on the Technical Basis for Revision of Regulatory Guide 1.99, consider that, with the current understanding of radiation damage mechanisms, it is not appropriate to use highly accelerated test reactor data directly to predict high-fluence behaviour for RPV or surveillance conditions. In contrast, other authors report that it is justified to use accelerated test reactor data directly to predict high-fluence behaviour for RPV or surveillance conditions, e.g. using irradiation data obtained from irradiations in the BR2 reactor (Chaouadi and Gérard, 2011) and the Halden reactor (Efsing et al., 2013).

The use of MTR data in evaluations of RPV materials from power reactors should be justified case by case (LONGLIFE recommendation). An example is given in the embrittlement assessment of Atucha I RPV (Wang, 2004) in which, due to the extreme low embrittlement rate of the MTR data observed for the Atucha I RPV material compared to that of the US power reactor data, further validation of the MTR accelerated data is required before it can be used for a life estimate of Atucha I.

Nevertheless, it should be mentioned that the combination of irradiation experiments in MTRs with modelling and microstructural studies provides an essential element in ageing evaluations of RPVs.

14. Integrated Surveillance Programme

The Integrated Surveillance Programme (ISP) is intended to substitute the existing plant surveillance capsule programmes with representative weld and base material data from host reactors. A representative material is a plate or weld material that is selected from among all the existing plant surveillance programmes to represent the corresponding limiting plate or weld material in a plant. The choice of a representative material considers chemistry (%Cu, %P, and %Ni), heat number, fabricator, heat treatment and welding process as it represents the plants' limiting materials. The best representative material is a material that has the following three qualities:

- A good or excellent chemistry match.
- The same welding process (if a weld), heat treatment and fabricator.
- Results in optimal consolidation of the test matrix (i.e., a candidate is better if it is capable of representing a number of plants rather than just one plant).

Even with materials from the same manufacturer, it is prudent to check that all the manufacturing processes are the same.

Two outstanding examples of Integrated Surveillance Programmes are the BWR Integrated Surveillance Programme of EPRI in the US (EPRI TR-1016575, 2012) and the Integrated Surveillance Programme for WWER-1000/V-320 in Eastern Europe (Brumovsky et al., 2005). The primary focus of the BWR ISP was to satisfy the requirements of 10CFR50 Appendix H for the BWR 40-year operating period. However, from the earliest stages of the ISP design process, it was recognized that the ISP test matrix could be logically extended to meet the needs of individual BWR utilities seeking plant life extension. The modified surveillance programme of the Temelin NPP with WWER-1000/V-320 reactor type is used for the ISP of several RPVs in Ukraine, Russia, Bulgaria and Czech Republic, as the standard surveillance programmes in WWER-1000/V-320 reactors do not fulfil all of the requirements of codes and standards (PN AE G-7-002-86) (PN AE G7-008-89) – e.g. non-uniformity of neutron field and even within individual specimen sets, large gradient in neutron flux between specimens and containers, lack of neutron monitors in most of containers and no suitable temperature monitors. Moreover, the location of surveillance specimens does not ensure conditions similar to those in the beltline region of reactor pressure vessels. In the modified surveillance programme of Temelin, irradiation of archive materials is performed together with the IAEA reference steel JRQ and a WWER-1000 reference steel. This allows more reliable and objective results to be obtained.

Minimum requirements for ISPs are given in Appendix H of 10CFR50 (10CFR50 App. H):

- The reactor in which the materials will be irradiated and the reactor for which the materials are being irradiated must have sufficiently similar design and operating features to permit accurate comparisons of the predicted amount of radiation damage.
- Each reactor must have an adequate dosimetry programme.
- There must be adequate arrangements for data sharing between plants.
- There must be a contingency plan to assure that the surveillance programme for each reactor will not be jeopardized by operation at reduced power level or by an extended outage of another reactor from which data are expected.

For life extension in particular, Section XI.M31 of the GALL report (NUREG-1801, 2010) recommends that Integrated Surveillance Programmes have a least one capsule with a projected neutron fluence equal to or exceeding the 60-year peak reactor vessel wall.

15. Coordinated Surveillance Programme

A Coordinated Reactor Vessel Surveillance Programme (CRVSP) can be used to obtain vessel embrittlement data with high fluence and long irradiation times. In a CRVSP several plants work together to optimize their specific surveillance programmes to support life extension to 60 or 80 years.

An outstanding example is the "Coordinated PWR Reactor Vessel Surveillance Programme", which is a joint effort of the PWR Owners Group and the Materials Reliability Programme of EPRI (Hardin, 2011a,b). This coordinated programme will generate the irradiated material samples and high-fluence surveillance data needed to support embrittlement correlation databases and damage mechanism assessments at fluences representative through 80 years of operation. Existing reactor vessel surveillance programmes of each USA PWR were reviewed (capsules contents, fluences, withdrawal schedules, etc.) and projected gaps in high-fluence data ($>3 \times 10^{19}$ n/cm^2) were identified. Changes to withdrawal schedules were recommended to obtain high fluence surveillance data for the full range of materials across the entire industry. The PWR CRVSP is intended to provide input to embrittlement trend curves with PWR data for use in 60 and 80 years RPV evaluations.

Important elements of any CRVSP are the following:

- Identify, for the reactors participating in the CRVSP, the range and average value of neutron fluence at 60 and 80 years.
- Group capsule materials based on product form (forging, plate, weld) and chemical composition (Cu, Ni, P, and Mn).
- Identify high fluence gaps for the different groups.
- Modify the management plan of the remaining capsules to yield high fluence Charpy data.
  Defer the withdrawal of selected capsules.
  Withdraw additional capsules not required in the original plan. e.g., use the standby capsules.
  Stagger withdrawals for plant of nominally identical manufacture to spread out the data.
- Support embrittlement trend curve development at high fluence.
- Maintain compliance with existing regulations and guidance (e.g. GALL report).

16. Microstructural analysis techniques

The extension of plant life from 40 to 60 years or 80 years requires beltline materials to operate under flux-fluence conditions for which there is currently little information. At the same time, the expansion of the beltline increases the range of materials exposed to significant radiation doses. Even with the information acquired within the existing research programmes worldwide, the amount of data available will be insufficient to allow secure extrapolation of current embrittlement trend curves to the new operational conditions. Mechanistic understanding of the degradation processes acting under these conditions is therefore essential to support the limited mechanical property data as it becomes available. Such understanding is most readily gained with the assistance of microstructural investigations.

On the basis of our current understanding, the RPV degradation processes most likely to operate at long times are hardening, grain boundary segregation and strain ageing. The first two lead to embrittlement, while the third affects the distribution of plasticity in different loading conditions. All three are associated with increases in the ductile-to-brittle transition temperature and reductions in the upper shelf. The hardening at long times is likely to be associated with the formation of:

- individual point defects;
- point defect clusters;
- including dislocation loops;
- Cu-rich precipitates;
- point defect-solute complexes;
- including segregated dislocation loops;
- P-rich precipitates;
- carbides and carbonitrides as well as
- Mn-Ni-Si-based precipitates.

The different rates at which these microstructural features develop affect the overall rate of hardening. Similarly, measurements of grain boundary composition can give independent information on the likely onset of grain boundary fracture. All the listed features are very small, and specialized experimental techniques are needed to acquire useful data on their development. None of these techniques require large specimens, so the addition of microstructural investigations to a surveillance programme does not require the diversion of much, if any space in a surveillance capsule. Small tiles of the order 10 mm × 10 mm × 1–2 mm may be added to the surveillance capsule if such space is available, but e.g. the regions of Charpy samples away from the fracture surface, or the heads of tensile specimens, are adequate to provide microstructural analysis specimens.

A list of the techniques most commonly used to investigate irradiated RPV steels is given in Table 2. For each of these techniques, careful analysis is required if the data are to yield useful mechanistic insight as to the development of the features observed.

Table 2
Microstructural analysis techniques.

Technique	Features observed	Comments
Positron annihilation, PA (including positron annihilation spectroscopy, PAS, positron lifetime measurements, PA-τ).	Vacancies, vacancy clusters, other defects which expand the lattice locally (e.g. interfaces, dislocations).	One of the few techniques capable of investigating point defects and their clusters (though not those of interstitial character). Chemical information can be ambiguous.
Transmission electron microscopy (including high-resolution TEM, scanning TEM, TEM with associated chemical analysis from X-ray or electron energy loss spectra).	Precipitates, dislocations, dislocation loops, grain boundary segregation.	Capable of investigating point defect clustering, but has a resolution limit of 1–2 nm for observation of features, higher for characterization.
Small angle neutron scattering, SANS.	Defect clusters consisting of solute atoms and/or vacancies (e.g. CRPs, MNPs).	Provides good statistical information on size distribution and indirect information on chemical composition of clusters.
Atom probe, field ion microscopy APFIM (including 3-dimensional instruments, TAP, 3DAP, PoSAP, EcoPoSAP, LEAP).	Solute distributions from compositional fluctuations to clusters and precipitates.	Modern instruments provide better statistics than older ones, but not as good as SANS. Nature of solutes involved clearer than with SANS, but fine details of spatial distribution affected by instrument character.
Auger electron spectroscopy (AES).	Grain boundary and surface compositions.	Requires intergranular fracture if grain boundary composition is to be observed. Semi-quantitative.
Scanning electron microscopy, SEM.	Structure of fracture surface.	Will show if deformation and fracture modes change with irradiation.

" PAS for irradiation and SANS for thermal stability"

Table 2 shows clearly that no one technique is capable of providing information on all the radiation-induced features, and small interstitial-based defects are particularly difficult to observe directly. Indirect observations with resistivity, conductivity, resistivity–Seebeck coefficient (RSC) measurements and Mossbauer spectroscopy have all been applied to irradiated material, but require even more care in their analysis, and intercomparisons, to avoid misinterpretation. Characterization of the defects according to their thermal stability, and comparing this with the thermal stability of hardening can be particularly helpful in determining the nature of the features observed and whether the features observed actually dominate the hardening process.

When the features are smaller than the resolution limit of the TEM, it is difficult to associate chemical information with physical information i.e. to determine if it is correct to analyze a particular SANS signal in terms of equivalent precipitates or extended regions of segregation along dislocations, whether the precipitates are homogeneously distributed or concentrated along grain boundaries (which would affect the resulting hardening), whether the solute clusters observed in the atom probe are independent or part of point-defect solute complexes, etc. While the radiation-induced features are very small, it is necessary to combine the results of several different techniques. A full investigation of the microstructure to complement the mechanical property measurements in a surveillance programme would involve at least PAS and SANS or AP (preferably at least one AP measurement to reduce the ambiguities in the PAS and SANS, or at least one SANS measurement to improve the statistics). It requires investigations of archive and as-irradiated material in more than one exposure condition in order to distinguish rates of development, together with a post-irradiation annealing sequence on one or more conditions to clarify correlations with hardening. AES becomes increasingly useful as SEM shows increasing amounts of intergranular fracture.

17. Conclusions

Monitoring embrittlement during life extension is a necessary task since there is not, in general, enough surveillance data available in the high fluence range. Different tools and techniques can be used to support embrittlement monitoring and evaluation during long term operation. Among the techniques are notably the reconstitution of broken specimens, the use of miniature specimens and the use of advanced fracture toughness approaches. The extension of the standard surveillance programmes can be performed in different ways, as for example by the irradiation of standby capsules or the movement of existing surveillance capsules to a higher lead factor position in the vessel. The design of new capsules is another possibility. It allows new instrumentation (dosimetry and temperature monitors) and new types of specimens to be introduced. Since there are still open issues in the high fluence range we are forced to act with prudence. In particular, the use of material test reactor data in power reactor evaluations should be justified case by case. Integrated and Coordinated Surveillance Programmes open the way to obtain vessel embrittlement data with high fluence and long irradiation times, and support the development of qualified embrittlement trend curves. Finally, it should be mentioned that microstructural analysis techniques are essential for mechanistic understanding of the degradation processes and for underpinning the mechanical property data.

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