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STUDIECENTRUM VOOR KERNENERGIE
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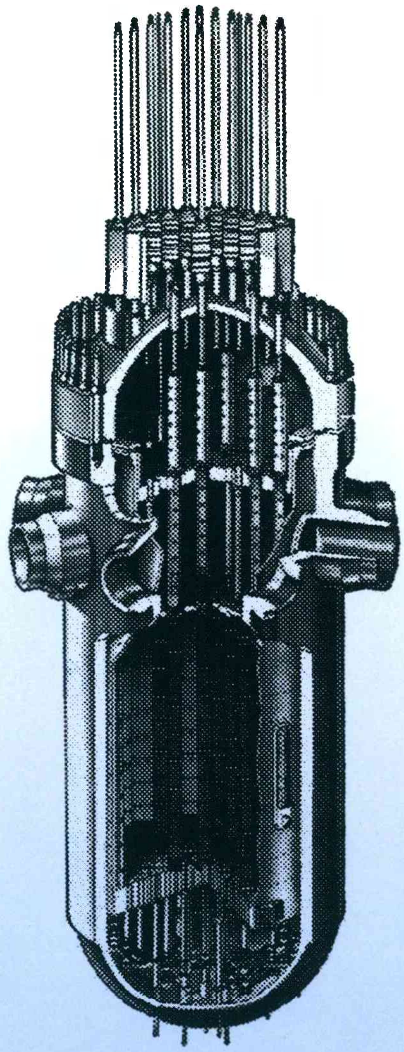
Topical Day – 18.03.1999

Ageing of Reactor Pressure Vessel Materials



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Introduction to RPV Life Management

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Nuclear Power Plant Life Management (PLIM) is a complex matter. As the reactor pressure vessel (RPV) is considered to be an irreplaceable component and as its safety against fracture has to be guaranteed at all times during the operational life of a nuclear power plant, RPV life management is a major safety issue. In fact, it is commonly said that the RPV Life determines the 'life' of the plant.

In PLIM terminology we have to distinguish between different 'lifetimes' that are primarily 'set' by the RPV life. We have the design life, the licensed life, the technical life and the economic life. PLIM encompasses the interplay between those different lifetimes to maximize the operational life of the NPP without compromising the safety of the plant.

The main actors in RPV management are the plant owner and operator (eventually its representative like the engineering office) and the legislator, or regulator. As is clear from below many times research institutes and international organizations are major players in the game to establish regulatory acceptance through their activities in the filed.

During plant operation the reactor pressure vessel – a low alloy steel structure with a brittle-ductile transition behavior – is at some 300°C (PWR-condition) and is continuously bombarded with neutrons. The combination of the neutron effects with possible effects from thermal ageing is a weakening of the material, so-called embrittlement. The evolution of the embrittlement is translated in a lowering of the fracture toughness – the resistance to crack development – of the material.
The implementation of reactor pressure vessel surveillance programmes that give anticipated information on the embrittlement of the vessel assure the safety of the pressure vessel. However, the safety evaluation depends on empirical rules, whereas the research and standardization community has proven that direct fracture toughness determination from small specimens, using an appropriate master curve methodology, is possible. This allows to get a realistic view on the fracture toughness of the material as a function of the received neutron fluence, and creates margins for improving plant life.
On the other hand, the research community tries to unravel the underlying phenomena that are responsible for the embrittlement behavior. Powerful microscopic techniques and thorough understanding of the metallurgical processes try to shed light on the damage mechanisms that occur into the material in order to predict the evolution of the fracture toughness of the material under NPP service conditions; again in view of margin reduction on existing legislation.

The Master Curve - From Idea to Standard

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The master curve method is a statistical, theoretical, micromechanism based, analysis method for fracture toughness in the ductile to brittle transition region. The method, originally developed at VTT Manufacturing Technology, simultaneously accounts for the scatter, size effects and temperature dependence of fracture toughness, as schematically presented in fig. 1.

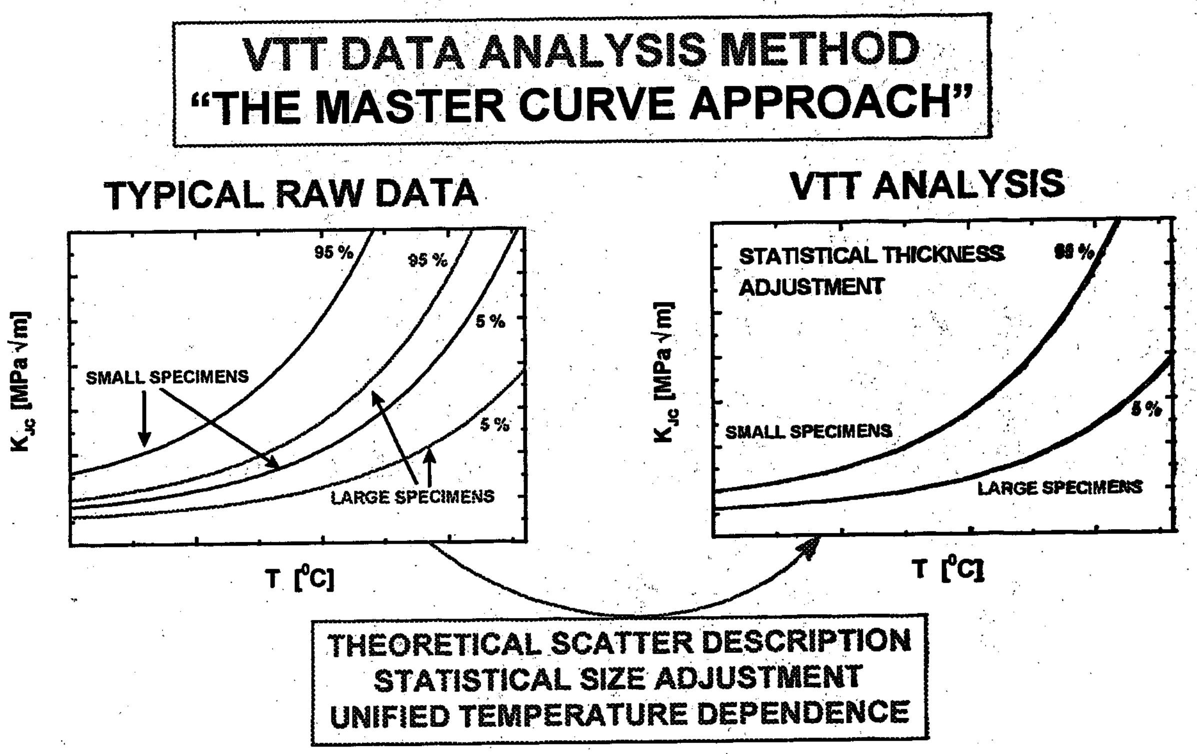

Fig. 1 Principle of the Master curve method.

The method has been successfully applied to a very large number of different ferritic steels and it forms the basis of a new ASTM testing standard for fracture toughness testing in the transition region. In USA, there is ongoing comprehensive validation and development work to include the master curve method into the ASME code as a new reference fracture toughness concept. Validation work is also ongoing within IAEA, the EC M&T programme MAT-CT-940080, the NFS project REFEREE, the Brite/Euram project SINTAP and the NESC programme.

The method enables the use of small specimens for quantitative fracture toughness estimation, thus reducing testing costs and enabling surveillance size specimens to be used for a direct measurement of fracture toughness. It also improves the quality of lower bound fracture toughness estimates, thus reducing the need for overly conservative safety factors. The applicability of the method is not restricted to nuclear applications. Its biggest impact is foreseen to be on fracture toughness determination for conventional structures, where testing costs and material use are presently inhibiting the use of fracture mechanics in design.

Master Curve Application to Cylindrical Geometry

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The Reactor Pressure Vessel (RPV) integrity is an important element in the safety against major accidental nuclear release. For this reason, the vessel design and operating sequences are based on a fracture mechanics approach. The mechanical properties of RPV steels are subject to degradation caused by neutron irradiation and thermal ageing. The embrittled material becomes more sensitive to cleavage fracture. Although failures of structures or components are seldom caused by cleavage fracture, cleavage is the most dangerous failure mode. Indeed, crack propagates instantaneously without prior deformation.

The properties of each RPV are monitored through the evaluation of tests on surveillance specimens loaded in surveillance capsules inside each reactor. Although some surveillance capsules contain fracture toughness specimens, the regulation is based on the transition temperature shift obtained by the Charpy impact test to evaluate the fracture behaviour through semi-empirical correlations, mainly based on a large experimental database. Sometimes, this semi-empirical approach, based on the Charpy specimen, results in a large conservatism penalising the operation of some reactors. Therefore, direct fracture toughness measurements on specimens are desirable.

However, fracture toughness determination in the transition regime suffers from very pronounced scatter, size effects and loss of constraint. The recent master curve approach is a powerful engineering tool to rationalise the size effect, the scatter and the temperature dependence of the fracture toughness in the transition region. In 1998, this method has been published as the American ASTM E1921 standard and is currently under consideration for standardisation in Europe. The master curve approach has also been used in a large experimental program (international Round Robin), which has confirmed its validity.

As the amount of irradiated material available is very limited, SCK•CEN thoroughly investigated the use of miniature specimen such as PreCracked Charpy V-notch (PCCv). However, this technique suffers from a reduced domain of applicability due to loss of constraint. In this context, a PhD work on the use of the small Circumferentially-Cracked Round Bar (CRB) was initiated at SCK•CEN. The presumed weaker size requirement is not the only advantage of the CRB. Other advantages are the low cost to machine the specimen, the rotating bending fatigue allowing easy precracking of specimens, the use of a standard tensile test fixture, the absence of shear lips, the possibility to load the specimen at very high loading rates and the axisymmetry of the specimen that avoids time-consuming 3D finite element calculations.

A large experimental program, featuring CRBs of different size and from well-characterised reference materials, in baseline and irradiated condition, was carried out at SCK•CEN. As the CRB is a non-standard geometry, the experimental program was supported by an in-depth theoretical study.

Analysing the experimental and theoretical results, we were forced to admit that the CRB also suffers from loss of constraint.

The existing procedure, which envisages censoring data subject to loss of constraint, would make the small CRB unusable. However, axisymmetric finite element calculations allows us to rationalise loss of constraint and to nevertheless apply the master curve approach.

Fracture toughness data from CRBs have been compared to larger standard bend geometry and to the small PCCv geometry. The following important results concerning the CRB were found:

- The temperature dependence of the fracture toughness is well represented by CRB data.
- The treatment of the size effect is applicable to the CRB.
- The scatter in the data can be rationalised by the Weibull statistic.
- The loss of constraint of CRB has been demonstrated and can be rationalised. This conclusion is reinforced by experiments performed on CRBs containing shallow cracks. The shallow crack is a well known configuration that amplifies loss of constraint effects.
- The CRB also allows to characterise material in its irradiated condition.

Besides validating of the master curve approach, this work enlightens the loss of constraint effect and suggests a way for rationalising it.

For completeness, the PhD thesis "Contribution to the Evaluation of the Circumferentially-Cracked Round Bar for Fracture Toughness Determination of Reactor Pressure Vessel Steels Toughness" will be available on the 25th of March, 1999 and will be publicly defended at the University of Liège in the month of May.

Microstructure Research in RPV Steel: Techniques versus Information

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The condition assessment of RPV's increasingly requires mechanistic understanding or framework. Particularly, when there is the required to extrapolate data to locations with
different dose, dose rates, uncertain or different material conditions, and a different or uncertain fabrication history. A requirement to develop and apply the necessary mechanistic understanding is proven experimental techniques to characterise the irradiation-produced microstructure. This presentation is concerned with demonstrating the need for such techniques, provide an overview of the current capability, and to illustrate the advantages of applying such techniques to RPV assessment.

The key to this approach is an agreed framework for the mechanisms controlling RPV embrittlement. There is a consensus that the fundamental degradation mechanisms are due to cluster hardening and non-hardening embrittlement. The cluster hardening leads to an increase in yield strength and an increase in the ductile to brittle transition temperature (DBTT), while irradiation-induced segregation of elements such as phosphorus to the grain boundary can lead to an increase in the DBTT but no increase in the yield strength.
The cluster hardening may be due to irradiation enhanced formation of copper-rich precipitates, and matrix damage due to radiation produced point defect clusters and dislocation loops.
The key requirements on microstructural techniques are to provide a full characterization of the irradiation-induced microstructure at or near the atomic scale. In particular to characterize the morphology, size and number density and composition of the small (< 2nm) copper-rich clusters, to quantify levels of embrittling elements in solution in the matrix and to characterize matrix damage, and to quantify the level of grain boundary segregants such as phosphorus and carbon.

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The current capability will be reviewed. Small angle neutron scattering (SANS), field emission electron microscopy (FEGSTEM) and atom probe (AP) now provide a proven means of fully characterising the size number density and composition of copper rich clusters. Although, there is the open question of how much Fe is in copper precipitates.The development of 3D AP (TAP, and OPoSAP) has significantly enhanced the power of the AP/FIM as a quantitative tool. Such studies have revived an early question of how much Fe is in the copper precipitates, which is an outstanding question.

Positron annhilation (PA) is now more routinely used, in particular in conjunction with post-irradiation annealing (PIA). Both lifetime and PALA studies have been reported.

Overall there has been greatest success in characterising Cu-rich precipitates with a well developed methodology for characterising number density, size and composition involving combination of techniques. There has been a consensus on development but some outstanding technical issues. In the case of matrix damage no technique has emerged which can directly resolve matrix damage clusters, but advances have been made into the nature of matrix damage and its parametric dependence by 'indirect observations' using positrons and post-irradiation annealing. In the case of grain boundary segregation techniques exist to measure levels of grain boundary coverage of important elements.

Illusrations will be given of the application of these techniques to irradiated RPV steels. Examples will be taken from studies which demonstrate the development of critical mechanistic insight which underpins RPV trend curve development. An important role for these microstructural techniques is to generate data which can be used to predict the change in macroscopic mechanical properties, in particular yield stress. Progress in this area will be discussed. Finally, the use of microstructural techniques to investigate outlier behaviour will be given.

Embrittlement of RPV-materials: Modelling Issues

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To date, safety assessment with respect to fracture of a RPV usually relies on an empirical or prescriptive approach. This works well if the design life of a new power plant has to be assessed. For the life extension of existing plants, however, many plant specific aspects concerning the operation conditions and their consequences on load and irradiation history determine the present state of RPV degradation and thus the plant integrity. Existing codes are not practicable for assessing the residual life of the plant under such conditions and more refined modelling approaches are necessary in order to avoid non- conservative or overconservative estimates. This requires a well-founded understanding of the basic mechanisms influencing materials embrittlement and the ductile-to-brittle transition (DBT) behaviour. Any conceptual breakthrough, especially with regard to the influence of microstructural mechanisms determining the DBT, may therefore have far-reaching practical consequences. Following this view, the theoretical concepts and microstructural mechanisms reviewed here may serve as a starting point for future modelling activities in order to achieve a physically sound assessment of the effects of thermal and irradiation-induced ageing on fracture mechanical properties, which is a central problem for ensuring long-term integrity of reactor pressure vessels.

This presentation aims to give an overview of the current theoretical understanding of the irradiation-induced materials embrittlement and to point out some fundamental yet unresolved questions, which deserve further theoretical investigations in the future. It is important to note that the study of these phenomena is involving researchers from different disciplines: solid state physics, physical metallurgy, mechanical engineering and nuclear engineering. On the one hand, this situation offers the possibility to combine top-down and bottom-up approaches in a multidisciplinary effort aimed to a more complete theoretical understanding of the ductile vs. brittle material behaviour in general. On the other hand, there is still some lack of communication between the different concepts used by the various disciplines. In this sense, an attempt is made to review physical and mechanical aspects of materials' embrittlement.

Various dimensional scales have to be taken into account for a comprehensive theoretical understanding of irradiation-induced embrittlement which correlate with the various experimental techniques available for qualifying embrittlement. First, some basic elements of the microstructural damage occurring during the formation of displacement cascades will be recalled. Then the irradiation-induced degradation of tensile properties, i.e. the hardening embrittlement due to the strengthening by matrix damage and precipitation will be discussed. On this level, important input to the theory comes from experimental techniques of microstructural characterization.

This results in a theoretical description of embrittlement in terms of a degradation of tensile properties, such as the yield stress and the strain-rate sensitivity which can easily be cross-checked experimentally.

The non-hardening component to embrittlement, i.e. a loss of ductility without concomitant strengthening of the grain matrix, is associated with the segregation of solute or impurity atoms to the grain boundaries, and acts upon the grain boundary cohesion strength.

This type of embrittlement is beyond the scope of the present review, because it comes along with a change in fracture mode from trans- to intergranular and because the factors controlling this are not yet as well understood as the hardening embrittlement.

The second part of the overview deals with the influence of tensile properties on fracture mechanics, i.e. with the materials behaviour in the presence of a crack. Various theoretical approaches have been proposed in the literature to model the DBT, which can roughly be classified as statistical continuum mechanics approaches based on the weakest link concept and deterministic approaches emphasising the role of the dynamics of dislocations in the plastic zone close to the crack tip. The principal ideas of these concepts will be outlined. Conclusions and a discussion of open questions will be given.

Dosimetry in RPVS: the Extra Dimension

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Since the reactor pressure vessel (RPV) is the primary containment of the nuclear reactor core, it is one of the most important safety components of a nuclear power plant. Radiation induced embrittlement of the RPV can limit the effective operating lifetime of the plant. Reactor PV integrity can also affect the cycle-to-cycle operations. The cycle-to-cycle limitations can arise since the normal strat-up and shutdown operating pressure-temperature (P-T) curves for a given power reactor can be influenced by the radiation-induced damage in the RPV.

In recognition of these safety and economic issues, the US NRC in association with various laboratories, among them ORNL, HEDL, NIST, SCK·CEN, started in 1977, the Light Water Reactor Pressure Vessel Surveillance Dosimetry Improvement Program (LWR-PV-SDIP) to improve, maintain and standardise neutron dosimetry, damage correlation, and the associated reactor analysis procedures used for predicting the integrated effect of neutron exposure to LWR-PV.

The major benefit of the LWR-PV-SDIP has been and continues to be a significant improvement in the accuracy of the assessment of the current exposure conditions of the RPV and the surveillance metallurgical samples allowing a better evaluation of the remaining safe operating lifetime of the RPV.

The extra dimension of the dosimetry rely on the fact that one needs to access the RPV data (neutron dose exposure, irradiation temperature) whereas the RPV can not be instrumented. Therefore one has to combine experimental data coming from the surveillance capsule located before the RPV and eventually from the ex-vessel dosimetry locations in the cavity behind the RPV with neutron transport calculation data.

To overcome this problem the dosimetry community have made an effort to access directly the experimental data of the RPV through scrapings to be taken from the inner face of the RPV and using the ^{93}Nb(n,n')^{93m}Nb reaction on the Nb impurities contained in the PV steel to obtain the fast neutron fluence accumulated in the PV. These kind of dosimetry measurement needs well established procedures and standards to ascertain the quality of the measured data.

Also the extra dimension of the dosimetry work, rely on the capability of the community to deliver the most accurate fluence values but with a well established precision and this is nowadays the challenge to answer for the community. Indeed, in assessing the uncertainty of the RPV fluence, it is important to remember that determination is not by C/E ratios, but must be assessed using all contributing error sources. Precision, may be found from experimental surveillance dosimetry results; 5% is a common value accepted nowadays. This lead to RPV fluence uncertainty values ranging between 15 to 20 %.

These values are nowadays agreed among the community thanks to the standardisation effort in reactor dosimetry, to the many benchmark experiments such as PCA, PSF, VENUS, NESDIP which revealed deficiencies such as the overestimation of the iron inelastic scattering cross-section, the perturbation effects of the detectors or their holders, or the photoreactions effects.

The last extra dimension and challenge for the reactor dosimetry community would be to make the fluence adjustment more physics based than a pure mathematical black box to make it acceptable by the regulatory bodies.

Life Management of Doel 1-2 Reactor Pressure Vessels

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Introduction

Commissioned in 1974 and 1975, Doel 1 and 2 were the first commercial nuclear power plants in Belgium. As in many other units of this vintage, the weld assembling the two core shells (forged rings) displays a relatively high copper content, detrimental to irradiation embrittlement sensitivity. Unusual irradiation embrittlement behaviour of this weld, coupled with a specific safety injection design, raised questions early on regarding the lifetime of the reactor pressure vessels.

The paper summarises the actions taken in the last 10 years by Tractebel Energy Engineering, in close co-operation with the Utility Electrabel and with the Belgian nuclear research center SCK•CEN, in order to successfully demonstrate that the integrity of the reactor pressure vessels (RPVs) could be justified for at least 40 years of operation.

Main Points

It is well known that Cu is one of the main elements responsible for the irradiation embrittlement sensitivity of RPV steels and welds. The Cu content of the Doel 1-2 weld surveillance specimens is generally in the range 0.12-0.15%, with the exception of specimens coming from a specific block whose Cu content reaches 0.35%. In both cases the Ni content is of the order of 0.15%. The first surveillance capsules showed a higher than predicted embrittlement of the low Cu welds, followed by an apparent saturation for the next capsules. Although all Charpy weld specimens come from the "low" Cu blocks, a reconstitution procedure developed and extensively validated at SCK•CEN made it possible to reconstitute irradiated high Cu Charpy specimens. Testing these high Cu specimens led to the unusual conclusion that the weld material irradiation embrittlement is practically independent of the Cu content.

Detailed characterisation of the composition of the irradiation-induced precipitates provided a possible explanation for this atypical behaviour. The densities and dimensions of the irradiation-induced precipitates are similar in the low and high copper weld materials, but their average composition is different. The precipitates in the low-Cu weld contain only 36% Cu and up to 35% Mn, while the average Cu content is above 70% in the high-Cu material.

The Pressurised Thermal Shock (PTS) loading was more severe in Doel 1-2 than in other units, due to the unusual direct safety injection in the downcomer (in addition to the injection in the cold legs). In order to mitigate this detrimental effect, a preheating of the safety injection water to 35°C was implemented, followed by more radical hardware modifications which eliminated the safety injection in the downcomer and redirected it to the upper plenum above the core. This modification made it possible to use the screening criteria from the U.S. regulation (10 CFR 50.61) to assess the risk of vessel failure in case of PTS. According to these criteria, the PTS risk remains acceptable, with margin, up to more than 40 years of operation.

Beyond this strictly regulatory approach, advanced techniques were applied in a defensive in-depth perspective, in order to show that the real embrittlement of the limiting weld material is much less severe than indicated by the regulatory approach.

The crack arrest fracture toughness curve K_{Ia} was evaluated by a novel approach developed at SCK•CEN, based on the arrest force measured in the instrumented Charpy tests. An "Arrest Force Master Curve" is used to derive an equivalent NDT, which is then used to index the ASME KIa curve. The initiation fracture toughness KIc was measured by means of pre-cracked Charpy specimens tested in three-point slow bending. The results were evaluated using the "Master Curve" approach, now standardised in ASTM E 1921. The comparison between the KIc fracture toughness shift and the RTNDT shift shows that the fracture toughness shift is apparently 10 °C larger, but this difference is not statistically significant considering the uncertainties. There is however a very large benefit on the irradiated fracture toughness curve due to the initial (unirradiated) condition, which is much better than indicated by the RTNDT-based approach. The difference between the unirradiated Master Curve and the regulatory curve is of the order of 80 °C.

Although the measured KIc and KIa curves may not be used in the regulatory justification at the present time, they demonstrate the conservatism of the regulatory approach and give increased confidence in the possibility to operate these units at least for 40 years, and probably longer. This will be confirmed as soon as surveillance capsules corresponding to a higher fluence are pulled out.

New PTS analyses, performed using the improved initiation and arrest toughness curves obtained with the advanced techniques, show that there is no crack initiation even for the most severe transient.

Conclusions This comprehensive approach, which extends well beyond the regulatory requirements, made it possible to demonstrate the integrity of the pressure vessels for 40 years of operation, and gives good reasons to believe that operation of these units beyond this limit will not be limited by the reactor pressure vessels.

=> Nancy

The Role of Metallurgy in RPV Steel Embrittlement

by
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Topical day on Ageing of Reactor Pressure Vessel Materials, Organised by SCK-CEN Mol, 18 March 1999

Synopsis

The various types of reactor pressure vessel (RPV) in use world-wide will be briefly described indicating the types of steels and welds used. The methods of RPV fabrication and the heat treatments employed will be indicated. The factors which affect the start-of-life properties will be outlined. The rules for operation of RPVs place certain requirements on the temperature and pressure margins available during RPV start-up and on-load operation which, in turn, has implications for the fracture toughness and strength of the materials used and their response at differing test temperatures. The limits are determined by the resistance to fast fracture of the most embrittled material and the resistance to deformation and plastic collapse of the weakest material. The issues involved in monitoring toughness degradation and strength changes using surveillance programme samples will be examined.

The modes of material property changes in service due to the effects of irradiation will be described. In general, the strength of a steel or weld is affected by matrix hardening arising from atom displacement damage and by matrix hardening due to the irradiation-induced precipitation of hardening phases. Both of these processes lead to embrittlement and a deterioration in the fracture resistance of the material concerned. There is also a third process of embrittlement that comes from degradation effects at grain boundaries; this arises from the segregation of certain elements that lower the grain boundary fracture strength. The general characteristics of these processes are described in terms of the influence of the main irradiation variables, namely neutron spectrum, dose, dose rate and irradiation temperature.

The role of fabrication variables, such as grain size, dislocation density and heat treatment, will be described together with the effects of chemical composition. These variable will be briefly considered in terms of their influence on the three embrittling mechanisms outlined above. The particular influences of C, N, B, Mn, Al, Si, Cu, Ni and P will be examined in relation to the possible underlying mechanistic understanding of the irradiation damage processes.

The possible methods for ameliorating the embrittlement of an existing vessel will be outlined and some of the principles involved in making improved vessels will be discussed.