REACTOR PRESSURE VESSEL EMBRITTLEMENT—I

Sponsored by the Radiation Protection and Shielding Division

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INTRODUCTION

The reactor pressure vessel (RPV) in a commercial nuclear reactor is expected to last the lifetime of the plant, a time in the range of 30 to 60 years. These vessels have traditionally been fabricated from a low alloy, Mo-Mn steel (e.g. ASTM A533, Grade B) in 6 to 8 in thick sections ranging. Both rolled plate and ring forgings have been used, with individual sections joined by submerged-arc welds. The RPV is a large cylinder on the order of 12 feet in diameter and 20 feet long. Among other functions, it serves as a primary barrier to fission product release in postulated severe accident scenarios, rendering in-service vessel failure unacceptable.

The steels used in manufacturing RPV steels have relatively high strength and good toughness in the as-fabricated condition. However, exposure to high energy neutrons leaking from the core leads to the formation of a fine-scale, radiation-induced microstructure that both further strengthens and embrittles the material. This embrittlement is observed as an increase in the ductile-to-brittle transition temperature (DBTT) which is characteristic of body-centered-cubic metals. At high neutron exposures, the DBTT can exceed room temperature, high enough to impact reactor operations and raise questions about RPV integrity under accident conditions.

Since the RPV is not a replaceable component, it became critical to understand the mechanisms that cause embrittlement. This information was used to develop mitigation strategies, and may provide better predictions of embrittlement in order to reduce the regulatory burden associated with uncertainties in material properties. Although the RPV is a large, low-technology engineering structure, the scientific challenge of developing an adequate understanding of their embrittlement has required application of both advanced microstructural characterization methods and computational simulations. These methods and their use are discussed below.

CHARACTERIZATION OF DEFECTS IN IRRADIATED RPV MATERIALS

Microstructural Investigations

Advanced techniques for microstructural characterization have been required to obtain an adequate description of the defects responsible for the increase in DBTT. Early metallurgical work indicated that the presence of low levels of copper (~10 to 30 wppm) as a tramp element increased the level of embrittlement [1]. Later, more detailed investigations revealed the impact of other minor solutes and alloying elements [2]. However, examination by transmission electron microscopy (TEM) revealed too few radiation-induced defects to account for the measured mechanical property changes.

The use of field ion microscopy in various forms, including atom probe tomography (APT) played a key role in identifying the defect responsible for the embrittlement, namely small copper-enriched precipitates or solute clusters. Several are shown in the APT three-dimensional atom map in Fig. 1 which reveals a high number density of Cu-, Mn- and Ni-enriched precipitates in this RPV weld that had been irradiated to a neutron fluence of $0.8 \times 10^{23}$ n. m$^{-2}$ (E $> 1$ MeV) at a temperature of 288°C. Both their small size and the fact that they are coherent with the bcc ferrite matrix limits their visibility under TEM.

Small angle neutron scattering (SANS) has also been heavily used in this research. SANS provides information about the evolution of copper-manganese-nickel precipitates as a function of key irradiation and environmental...
variables. Fig. 2 shows the evolution of mean precipitate radius with increasing fluence in two A533B model that contain 0.4% Cu, 0.8 and 1.25% Ni and 1.4% Mn for the circles and squares, respectively. Combined with the information on number density and volume fraction that is also obtained, the results indicate that the precipitates undergo a slow coarsening mediated growth with increasing fluence.

**Computer Simulation**

Atomistic simulation methods, including molecular dynamics (MD) and Monte Carlo (MC), have been used to investigate the formation and evolution of the radiation-induced defects. MD simulations displacement cascades of have provided a detailed description of primary damage formation [3]. Various MC methods have been used to both age the residual defect distributions obtained from the MD, and to investigate the clustering of solutes in an irradiated material. A typical result from the latter work is shown in Fig. 3. This figure shows a snapshot of an approximately 1.6 nm radius precipitate with a composition of 60% Cu, 5% Ni, 5% Si and 19% Mn. The Monte Carlo predictions are in good agreement with APT measurements that indicate enrichment of Mn and Ni at the precipitate/matrix interface.

MD simulations of the interaction between dislocations and radiation-produced defects have been used to investigate the details of hardening mechanisms. The information obtained from the atomistic simulations is being applied in kinetic models to predict the fluence and temperature dependence of embrittlement.

**SUMMARY**

Atomistic and mesoscale computer simulations and high resolution microstructural characterization have provided the basis for understanding the underlying physical mechanisms of radiation-induced embrittlement in RPV steels. This information is having an impact on reactor operations and regulatory decision making.

**REFERENCES**

INTRODUCTION

The main activities of WWER dosimetry arise from necessity of solution of the following actual problems:
- reliable assessment of reactor pressure vessel embrittlement for WWER-440 and WWER-1000 reactors,
- prolongation of the RPV lifetime for WWER-440s beyond designed terms.

The improvement of the reactor pressure vessel (RPV) dosimetry in WWER type reactors leads to the multifactor approach taking into account not only the fast neutron fluence [1]. The increase of the precision of the description of the mixed photon and neutron radiation field needs careful benchmarking to eliminate the probable biases.

Three models (mock-ups) of WWER type reactors assembled in the LR-0 experimental reactor in NRI (Řež, Czech Republic) were qualified [2] for benchmarking of RPV dosimetry in the frame of a European Project REDOS [3]. Radial full-scale mock-ups, which copy geometry and material composition of the core periphery, reactor internals, RPV, and biological shielding, represent the engineering benchmarks. The mock-ups are azimuthally (60 degrees) and axially (1.25 m fuel active length) shortened. The mock-ups were designed with respect to possibility to provide the best guess of the space-energy distribution of fast neutrons for the RPV monitoring system, thus the operational density of moderator was simulated in downcomer by insertion of an air layer (displacer).

RECENT ACTIVITY IN BENCHMARKING

The qualified set of LR-0 benchmarks consists of WWER-440 with “standard” core loading, WWER-440 with dummy steel assemblies at core periphery, and WWER-1000 with simple core loading (mostly with uniform enrichment). The neutron spectra measured at the barrel and over RPV (with the step 5 cm) in the energy range 0.1-10 MeV provide a valuable test of calculation models, methods and data libraries. The measurements were carried out in the central plane of mock-up along the mock-up axis of symmetry.

The gamma radiation space-energy distribution has been measured recently over the RPV thickness in the WWER-1000 mock-up [4]. The improved spectrometer (multiparameter analysis) with stilben scintillator [5] enables simultaneous measurement of fast neutron and gamma spectra.

All measurements are accompanied by calculations (deterministic and Monte Carlo).

RESULTS

Some experimental integral data obtained in the WWER-1000 mock-up are demonstrated in the Table 1. The scheme of the WWER-1000 mock-up as well as short review of the neutron measurements and calculations can be found e.g. in [2,6]. The RPV consists of four 5 cm thick layers, which can be moved relating to the detector position. The points of measurement on the surfaces and over the RPV are marked in following way: point 3 – before RPV, 4 – 1/4 of RPV, 5 – 1/2 of RPV, 6 – 3/4 of RPV, 7 – behind the RPV. N(E,i) and G(E,i) in the Table 1 mean integral neutron and gamma fluxes above energy threshold E, MeV, in point i. Calculated data in the Table 1 were obtained with Monte Carlo code MCU-REA/2 and nuclear data library DLC/MCUDAT-2.2. This code was developed starting from universal Monte Carlo code MCU-RFFI [7] for WWER core and out-of-core calculations.

All relative distributions are normalized to 1 in point 7, behind the RPV (mandatory point of neutron monitoring in the WWER NPP). The spectral indices N(0.5,i)/N(3,i) and N(1,i)/N(3,i) demonstrate the softening of the neutron spectrum over the RPV thickness. On the contrary the gamma spectra have the same shape except the point 3 with predominant contribution of 2.23 MeV photons from Hydrogen.

The possibility to recognize the energy structure of the gamma spectrum is demonstrated in Fig. 1.
The spectra measurements were repeated several times in each point and the averaged ones have been used for further evaluation. The sigma of these averaged values varies from 1 to 10% in energy groups and the sigma of integral values is correspondingly reduced. The long time stability of the LR-0 power monitors is 2%.

REFERENCES


Analysis of Irradiation Conditions of WWER-1000 Surveillance Specimens

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INTRODUCTION

Surveillance specimens are necessary for reliable evaluation and prognosis of reactor pressure vessel (RPV) embrittlement and lifetime. These goals can be reached only under reliable dosimetry of surveillance specimens, especially in cases of complicated design and location of specimen capsules. It is a specific situation of WWER-1000 surveillance program. Location of surveillance assemblies in a reactor is shown in Fig. 1, locations of containers inside assembly – in Fig. 2.

Some features of the WWER-1000 surveillance program and concerned dosimetry problems and difficulties were discussed in [1]. In particular, it was demonstrated, how the more precise dosimetry of WWER-1000 surveillance specimens can lead to significant changes in an evaluation of radiation embrittlement of RPV materials.

EXPERIMENTS AND CALCULATIONS

The first special experimental – calculation investigation of the neutron field in WWER-1000 surveillance assemblies was carried out under TACIS Project R2.06/96 [2]. Two experimental surveillance assemblies (full copies of standard ones) equipped with $^{93}\text{Nb}(n,n')$, $^{54}\text{Fe}(n,p)$, $^{58}\text{Ni}(n,p)$, $^{46}\text{Ti}(n,p)$, $^{63}\text{Cu}(n,\alpha)$ activation monitors were located in the standard positions on the baffle of Balakovo Unit 1 WWER-1000 and irradiated during one fuel cycle. The locations of activation monitor sets are shown in Fig. 2. The monitors were located inside notches of Charpy specimens and outside specimens.

Fig.1. Location of a Surveillance Assembly in a WWER-1000 Reactor

Fig.2. Location of the Monitor Sets in the Experimental Surveillance Assemblies (top view)

The errors of measured end-of-irradiation monitor activities was evaluated as (2-6)% (1 sigma).

The first calculations were deterministic [2]. The second series of calculations were carried out with Monte Carlo code MCU-REA/2 and nuclear data library DLC/MCUDAT-2.2. The calculations included two steps. In the first step the source for the second one was accumulated on the surface of specimen assembly. Due to relatively small value of assembly surface (in comparison with reactor dimensions) the nonanalog modeling using weight windows method was used in the first step. The calculated activities of relatively short-lived $^{58}\text{Ni}(n,p)$ and $^{46}\text{Ti}(n,p)$ monitors were corrected taking into account the neutron field changes in the upper part of the core during fuel cycle.
RESULTS

General comparison of experimental and Monte Carlo results is shown in the Table 1, where data averaged over all monitor sets in both assemblies are included. The dispersion of Monte Carlo calculation depends on assembly, monitor position (due to different distance from the core), and energy threshold of monitor reaction. The overall dispersion limits are shown in the Table 1 for each monitor.

Average agreement of experiment and calculation is more or less well for all monitors except $^{63}\text{Cu}$, although the spread of discrepancies seems to be too high, so the local analysis should be continued. Discrepancies of copper monitor activities partly seem to be caused by poor statistics in calculations.

<table>
<thead>
<tr>
<th>Monitor</th>
<th>Effective threshold, MeV</th>
<th>Dispersion limits of MCU calculations, %</th>
<th>Average ratio MCU/Exp</th>
<th>Standard deviation of MCU/Exp, %</th>
<th>Maximal ratio MCU/Exp</th>
<th>Minimal ratio MCU/Exp</th>
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<td>$^{93}\text{Nb}$</td>
<td>0.5</td>
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<td>$^{54}\text{Fe}$</td>
<td>2.5</td>
<td>2.0-10</td>
<td>1.01</td>
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<td>2.0-7.0</td>
<td>0.98</td>
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<tr>
<td>$^{46}\text{Ti}$</td>
<td>4.6</td>
<td>3.0-14</td>
<td>0.92</td>
<td>5.0</td>
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<td>$^{63}\text{Cu}$</td>
<td>7.0</td>
<td>4.0-23</td>
<td>0.87</td>
<td>8.4</td>
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REFERENCES


INTRODUCTION

The current methodology for the determination of the fast fluence \((E > 1 \text{ MeV})\) inside the reactor pressure vessel (PV) wall, acceptable to the U.S. Nuclear Regulatory Commission (NRC), is described in Regulatory Guide 1.99, Revision 2.\cite{1} The regulatory guide states that “The neutron fluence at any depth in the vessel wall, \(f \left(10^{19} \text{n/cm}^2, E > 1 \text{ MeV}\right)\), is determined as follows:

\[
f = f_{\text{surf}} \times e^{0.24x}
\]

where \(f_{\text{surf}} \left(10^{19} \text{n/cm}^2, E > 1 \text{ MeV}\right)\) is the calculated value of the neutron fluence at the inner wetted surface of the vessel at the location of the postulated defect, and \(x\) (in inches) is the depth into the vessel wall measured from the vessel inner (wetted) surface. Alternatively, if dpa calculations are made as part of the fluence analysis, the ratio of the dpa at the depth in question to dpa at the inner surface may be substituted for the exponential attenuation factor” in Equation 1.

The attenuation formula (1) was constructed to match the average dpa attenuation rate observed in Guthrie’s calculations of six different PWR reactors.\cite{2} Since Guthrie’s analysis, the cross sections were updated several times. A \(P_3\) expansion is routinely used in the transport calculations (while Guthrie used \(P_1\)), and the cross-section libraries typically use finer energy groups. Given all these changes, it appears necessary to revisit the fast flux and dpa attenuation in the PV wall, which is the purpose of the work presented here.

ANALYSIS AND RESULTS

All neutron transport calculations were performed with the DORT computer code. The synthesis method, which combines one \(r-\theta\), one \(r-z\), and one one-dimensional \(r-\) calculation to obtain the three-dimensional flux distributions, was used throughout the analysis. All calculations used a \(P_3\) expansion of the scattering cross sections, except those calculations in which the effect of the order of scattering expansion was investigated. The fixed neutron source spatial distributions were prepared from the core power distributions obtained from the fuel cycle calculations, which were not part of this study, and were performed with codes used for core design. Two neutron cross-section libraries were used. The BUGLE-96 library, released in 1996, is the currently recommended cross-section library based on ENDF/B-VI.\cite{3} The SAILOR library represents older cross-section data and is based on ENDF/B-IV.\cite{4} The calculations with the SAILOR library were done to quantify the change in the flux and dpa rate attenuation in the PV wall due to the change from ENDF/B-IV to ENDF/B-VI cross sections.

The initial calculations were done with the models based on the as-built data for the H. B. Robinson-2 (HBR-2) three-loop pressurized-water reactor (PWR) of Westinghouse design, with a reactor thermal power of 2300 MW. In the subsequent calculations the thermal shield and PV wall thicknesses were varied to assess the effects on the attenuation in the PV. Finally, the analysis was also performed for an 1876-MW (thermal power) two-loop PWR of Westinghouse design. The attenuation curves for the two-loop plant (with PV thickness of 17.16 cm) closely agreed with the results for the three-loop plant with similar PV thickness (i.e. 18.75 cm).

The study is documented in [5]. Examples of the results are shown in Fig. 1.

CONCLUSIONS

The attenuation of the dpa rate in the PV wall calculated with the BUGLE-96 cross-section library, a \(P_3\) approximation to the angular dependence of the scattering cross sections, and dpa cross sections derived from ENDF/B-VI data is significantly slower than the attenuation predicted by the RG 1.99, Rev. 2, formula. For a PV wall thickness of ~24 cm, the calculated ratio of the dpa rate at 1/4 and 3/4 of the PV wall thickness, to the dpa value on the inner PV surface is ~14%, and 19% higher, respectively, than predicted by the RG 1.99, Rev. 2, formula.

The dpa rate attenuation in the PV wall, calculated with the SAILOR (ENDF/B-VI) cross-section library, a \(P_1\) approximation to the angular dependence of scattering cross sections,
Fig. 1. Flux (E > 1 MeV) (top) and dpa rate (bottom) attenuation in the PV wall, calculated with BUGLE-96 library and the ENDF/B-VI dpa cross sections, for different PV wall thicknesses.

and dpa cross sections from ASTM standard E 693, comes relatively close to the RG 1.99, Rev. 2, formula prediction. The calculation still predicts up to ~5% slower dpa attenuation in the region close to the PV inner wall.

Insight into the causes of the large difference between the dpa attenuation in the PV wall obtained with the “BUGLE-96, P3, ENDF/B-VI dpa” calculation and the RG 1.99, Rev. 2, formula was obtained from the comparisons of results with the BUGLE-96 and the SAILOR cross-section libraries, the $P_1$–$P_3$ approximations for angular distributions of scattering cross sections, and the ASTM and the ENDF/B-VI dpa cross sections. Approximately 40–45% of the difference is caused by the change in cross sections (from SAILOR, $P_1$ to BUGLE-96, $P_1$), ~35–40% of the difference is due to the change from $P_1$ to $P_3$, and ~15–20% of the difference is caused by the change from the ENDF/B-IV to the ENDF/B-VI dpa cross sections. Near the PV surfaces the calculated attenuation of the dpa rate also deviates from the simple exponential attenuation used in the RG 1.99, Rev. 2, formula, which contributes to the larger differences, especially near the PV outer surface.

REFERENCES

RPV Integrity Assessment by Operational Feedback: Post Service Investigations of VVER-type NPPs

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INTRODUCTION

Pressure vessel integrity assessment after long-term service irradiation is commonly based on surveillance program results. Radiation loading, metallurgical and environmental histories, however, can differ between surveillance and RPV materials. Therefore, the investigation of RPV material from decommissioned NPPs offers the unique opportunity to evaluate the real toughness response. This has been done several times, but only at prototype reactors. A chance is given now through the investigation of material from the decommissioned VVER-type Greifswald NPP to evaluate the state of a standard RPV design and to assess the quality of prediction rules and assessment tools.

CHARACTERISTICS OF THE GREIFSWALD NPP

Between 1973 and 1979, four units (unit power 440 MWel) of the Russian Pressurized Water Reactor line VVER-440/230 were put into operation in Greifswald (East Germany). Design, material production and operation conditions are identical or almost identical for more than 30 similar units in Russia and Eastern Europe. Their RPV material is very suitable for investigating embrittlement phenomena: The neutron flux is very high (small water gap: 16 cm), the operation temperature is low (270 °C) and the material is sensitive to neutron embrittlement (Cu content > 0.12 %, high P content in weld). Therefore the accepted embrittlement limits were already reached after about 12 cycles, and the units had to be annealed around the most critical weld 4 (see below) to continue their operation. In 1990, after the German reunification all units were shut down.

The dismantling of the unit 1 RPV will start this year. The RPV will be moved to a special cutting place, where it will be horizontally cut in rings (approx. 1 m high). Then the rings will be cut by a vertical saw in pieces (approx. 40 cm broad). For the material investigation program pieces of 125 x 125 mm with a thickness of 140 mm will be cut with the same system. It was proven by test cuts that the temperature increase during cutting is insignificant. The temperature was measured by thermocouples and an infrared camera.

INVESTIGATION PROGRAM AND RESULTS

Fluence Calculations

Knowledge of neutron fluence integrals \( \phi_{E>0.5\text{MeV}} \) and \( \phi_{E>1\text{MeV}} \) and dpa-values at the inner and outer pressure vessel walls are prerequisites for the material investigations. They were obtained for all four units for 10 height layers and 3° azimuthal intervals with the help of a Green’s functions method. The Green’s functions were calculated earlier by the Monte Carlo code TRAMO [1]. Some of these results characterizing the irradiation state of the Greifswald units are included in Table 1.

Further calculations will include studies of the influence of gamma irradiation, of the radiation spectra and of flux effects. Time dependent neutron and gamma flux spectra will be calculated. Fortunately, a very detailed documentation exists of all needed reactor data over the whole time of operation of each unit. Based on new calculations of pin-wise time-dependent neutron sources and a modernized nuclear data base, Monte Carlo calculations are being prepared of time-dependent neutron and gamma fluence spectra at all positions throughout the RPV wall, where the material pieces will be taken.

Material Investigations

The working program is focused on the characterization of the material state. It comprises chemical analysis, microstructure investigations (by means of metallography,
electron microscopy and SANS) and mechanical testing (hardness measurements, tensile tests, Charpy tests, fracture mechanics tests). The material investigation will be carried out based on pieces from the following 4 height positions (in mm relative to the core midplane, core height 2500 mm):

- Reference material position (weld 3) at -2825 mm (below core)
- Critical weld position (weld 4) at -915 mm
- Maximum neutron load position (base metal) at 0 mm
- Reference material position (weld 5) for estimation of thermal aging and gamma effects at +1715 mm.

The following results of the program can be expected [2]:

- Determination of fracture toughness parameters at irradiated conditions \( T_0, K_{IC} \)
- Characterization of spatial variability and homogeneity of real vessel wall and weld properties
- Verification of toughness parameters correlations
- Validation of code approaches and embrittlement predictions
- Validation of plant annealing as industrial process
- Evaluation of the re-embrittlement behavior
- Identification of the most critical safety-related issues
- Enlargement of the data base.

Simultaneously, material stocks for further research programs including benchmark experiments to verify NDT embrittlement characterization methods will be obtained.

**REFERENCES**

2. Tacis-project NUCRUS 96601, Final Report, Brussels 2000

<table>
<thead>
<tr>
<th>unit</th>
<th>cycles</th>
<th>Effective days</th>
<th>An-nealed in</th>
<th>Azimuthal maximum of ( \phi_{E&gt;1MeV} ) in units of ( 10^{19} \text{n/cm}^2 )</th>
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*after annealing
Pressure Vessel Enhanced Surveillance at the Argentine Heavy Water Reactor Atucha 1

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INTRODUCTION

The Argentinean nuclear power plant Atucha-1 (CNA-1) is unique in its type. CNA-1 is a 370 MWe pressurized heavy water reactor (PHWR) that employs low enriched uranium (LEU – 0.85 % $^{235}$U) as fuel and heavy water as moderator, reflector and coolant. CNA-1 has basic features of its own, like a reactor pressure vessel (RPV) 12 m high, 5 m in diameter and 22 cm thick, made of 22 NiMoCr 3 7 ferritic steel, and operating at 265 °C with an internal pressure of 121 atm.

Embrittlement predictions based on well-established LWR trend curves cannot be used directly in the case of CNA-1 RPV. Differences in coolant temperature and neutron spectrum introduce uncertainties in the prediction of the CNA-1 vessel embrittlement and penalize its operating margin.

CNA-1 RPV SURVEILLANCE PROGRAM

Over decades the CNA-1 RPV Surveillance Program, implemented in 1974 when the plant started operation, has been providing data to continuously monitor the irradiation embrittlement of the CNA-1 RPV material. However, as a consequence of the reactor design, the surveillance samples were located at the only accessible position, i.e. attached to the fuel element channel nozzle. So, our present knowledge of the CNA-1 vessel embrittlement is based on the results of specimens irradiated at below-core positions where the neutron spectrum is dominated by a large thermal neutron component. This makes it difficult to compare CNA-1 surveillance results against international databases or against the complementary irradiations that were carried out in 1983 at the German reactor VAK.

Recently, the embrittlement results obtained at VAK under a typical PWR neutron spectrum could be successfully correlated with those obtained at CNA-1 utilizing surviving defects as neutron exposure parameter [1]. Based on these preliminary results a re-evaluation of the surveillance database was carried-out that lead to satisfactory conclusions concerning the vessel integrity.

CNA-1 RPV Enhanced Surveillance Program

To confirm these results, extend our surveillance database, and prepare the future RPV surveillance beyond design life an enhanced surveillance program was launched in November 2002 at the Belgian Nuclear Research Centre (SCK•CEN), Mol, Belgium. Within the Belgo-Argentine Bilateral Cooperation Agreement a project defining 3 parallel tracks was signed: The interrelated issues of Mechanical Testing and Dosimetry, together with modeling in support to the safety evaluation of the CNA-1 vessel [2].

NEW IRRADIATIONS AT BR2 BELGIUM

Irradiations of CNA-1 archive material are due to start next July at the Belgium reactor BR2. The CNA-1 specimens will be irradiated together with reference material JRQ at the irradiation channels IPS1 and IPS3 of the Callisto-Loop where the irradiation temperature matches that of the CNA-1 RPV wall. The irradiation will be conducted in one cycle, lasting 28 days. Two fluences will be targeted, one close to the EOL fast fluence; the second one will be beyond the EOL to check the steel behavior for safe operation of the RPV.

Preliminary results show that some spectral differences appear when comparing the neutron flux at position IPS3 of the BR2 reactor [3] and at the inner surface (IS) of the CNA-1 RPV wall [4], see Fig. 1. The neutron flux spectrum at IS was obtained using the 3D discrete-ordinate transport-code TORT in R-$\theta$-Z geometry and $P_3$S$_8$ approximation. Cross-sections were taken from BUGLE-96 library in 47-neutron energy group structure. The spectrum at the Callisto channel IPS3 was calculated using the Monte Carlo code MCNP-4 with cross-sections derived from file ENDF/B-6.
Fig. 1: Percent contribution to the total flux at irradiation position IPS3 of the Callisto-Loop and in capsule BA1 of the VAK reactor, as compared to that found at the inner surface (IS) of the CNA-1 RPV.

These preliminary results show that neutrons with energies above 0.1 MeV contribute with 8% to the total flux at IS while this contribution is higher at IPS3 (~25%) and at the capsule BA1 irradiated at VAK (~22%). Also the thermal groups contribution to the total flux amounts to 26% for IS and is higher for both IPS3 and BA1 cases. Therefore, a thorough analysis of spectral effects on defect production is needed if the results of mechanical tests are going to be correlated with the actual vessel damage.

Further modeling work, improvement of the calculations methods, and extension to embrittlement correlations using surviving defects as neutron exposure parameter, are foreseen.

REFERENCES

A New Methodology for Developing Charpy Impact Data Trend Curves

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INTRODUCTION

A new methodology that incorporates the chemical compositions into the Charpy trend curve was developed. The purpose of this new fitting procedure is to generate a new multi-space topography that can properly reflect the inhomogeneity of the surveillance materials, and utilize this multi-space trend surface to link and to project the surveillance test results to that of reactor pressure vessel steels.

SURVEILLANCE CHARPY TEST DATA

Currently, the Charpy impact test data are widely used by industry and regulatory agencies to monitor the reactor pressure vessel (RPV) degradation during service. In general, the impact energy of the Charpy impact test samples from RPV surveillance capsules depends on irradiation temperature (T), neutron fluence, and chemical compositions and material history of the test samples, which can be described as below.

\[ E = f(T, \varphi, Cu, Ni, Microstructure,...) \]

Mean fluence and irradiation time are generally used for data from the same capsule; and RPV materials, such as base, weld and HAZ, are categorized within separate groups; thus, the impact of microstructure factor can be minimized. Therefore, the formulation of the impact energy for a Charpy impact data set from one surveillance capsule can be further simplified and written as follows:

\[ E = f(T, Cu, Ni, ...) \]

Proposed New Fitting Function with Consideration of the Chemistry Variability

In order to consider the chemical variability of the surveillance test samples, a new fitting procedure that incorporates the chemical composition into the governing equation was developed. The formula for the impact energy as a function of test temperature (T), and copper and nickel (or plus other chemistry, such as P, Mn, etc.) can be written as follows:

\[ E = \left\{ A + B \cdot \text{Tanh} \left[ \frac{T-T_0}{C/f_2} \right] \right\} \cdot f_3 + f_4 \]

where \( f_i, i = 1,4 \) are functions of chemical composition for Cu and Ni content, and \( A, B, C, \) and \( T_0 \) are fitting parameters. This new fitting procedure provides a new multi-dimension topography that can properly reflect the inhomogeneity of the surveillance materials.

Results of the Proposed Fitting Procedures

A FORTRAN program was written for this study, using the IMSL non-linear-least-squares fitting subroutine ZXSSQ with three convergence criteria specified, including the difference of residual sum of squares from two successive iterations is within the specified limit. The residual is defined as the measurement minus prediction.

The weld Charpy data, with heat_id = WDR301 listed in the Embrittlement Data Base (EDB) [1], from surveillance capsule 18 of Dresden Unit 3 nuclear power plant were used for this feasibility study. The new fitting procedure achieves a reduction in uncertainty of predicted impact energy by 27%, compared to that of the conventional hyperbolic tangent fit procedure, which are illustrated in Figs. 1.

CONCLUSION

Based on the constructed multi-space topography, as illustrated in Fig. 1, one can substitute the target chemistry into integrated formulation of the chemistry composition functions, \( f_i \).

REFERENCES

INTRODUCTION

The discrete ordinates (S\textsubscript{N}) method [1] is commonly used in solving the neutron transport equation for pressure vessel dosimetry applications. In the S\textsubscript{N} method, the energy dependency is represented by the multigroup methodology, which is based on a three-step procedure, as follows [2]:

Step 1) Point-wise cross sections are processed to obtain fine-group cross sections.

Step 2) Self-shielding calculations are performed.

Step 3) Fine-group cross sections are collapsed into broad groups.

One of the important factors that affect the accuracy of multigroup libraries is the group structure. Although there are widely used multigroup libraries available for specific applications, there is no publicly available group construction methodology that can effectively develop group structures for a problem and/or objectives of interest. Hence, to increase the accuracy of multigroup libraries, we have developed a new, iterative, problem- and objective-dependent group structure construction methodology, referred to as the CPXSD (Contribution and Point-wise Cross Section Driven) methodology [3]. In this summary, we briefly introduce the CPXSD methodology, generate new libraries for a pressure vessel problem, and benchmark these libraries.

APPLICATION OF THE CPXSD METHODOLOGY

To test the effectiveness of the CPXSD methodology, we constructed fine- and broad-group libraries for pressure vessel studies using the Three Mile Island Unit-1 (TMI-1) reactor. In constructing group structures, we concentrated on both fast (i.e., above 0.1MeV) and thermal energy ranges (i.e., below 5eV) for fast neutron dosimetry and gamma production from thermal neutrons, respectively. For fast neutron dosimetry we considered six dosimetry isotopes including the \textsuperscript{63}Cu(n,\alpha), \textsuperscript{54}Fe(n,p), \textsuperscript{58}Ni(n,p), \textsuperscript{46}Ti(n,p), \textsuperscript{237}Np(n,f) and \textsuperscript{238}U(n,f) reactions at the cavity dosimeter. For gamma production from thermal neutrons, we considered the \textsuperscript{1}H(n,\gamma) and \textsuperscript{56}Fe(n,\gamma) reactions in the downcomer, and pressure vessel, respectively.

DISCUSSION ON THE CPXSD METHODOLOGY

The CPXSD methodology constructs fine- and broad-group structures considering two criteria: i) importance of groups and ii) point-wise cross sections of an isotope/material of interest. The importance of groups can be determined considering the group-dependent response flux formulation [4] given by:

\[
C_g = \sum_{i \in D} V_i \sum_{l=0}^{i} \sum_{m=0}^{l} \frac{2l + 1}{4\pi} \psi_{l,m}(\hat{r}, \hat{s}) \psi^*(l,m; \hat{r}, \hat{s}),
\]

In Equation (1), \(V_i\) is the volume of the sub-domain, \(l\) and \(m\) are azimuthal and polar indices, \(g\) is group, \(\psi\) is the angular flux, and \(\psi^*\) is the adjoint function.

The CPXSD methodology constructs group structures by refining an initial arbitrary group structure, considering the two aforementioned criteria. First, the objectives are calculated using the cross-section library having the initial group structure. The importance of groups is calculated and the most important group is identified. Depending on the point-wise cross sections of the important isotope/material, sub-groups are placed in the most important group, considering the resonance and non-resonance behavior of cross sections. After the number of sub-divisions in the most important group is obtained, the number of sub-divisions in other groups are determined based on the ratio of their \(C_g\), to the maximum \(C_g\). Sub-divisions in other groups are performed and a new group structure is generated. The refinement process continues until a convergence criterion on the objectives is achieved.
epithermal to thermal groups for the epithermal energy range, and reaction rates per unit energy and transfers within thermal groups for the thermal energy range. We selected $^{56}\text{Fe}$ as the important isotope in the problem and used its point-wise cross sections for determining the boundaries of the sub-groups. Then, the CPXSD methodology was used to construct new group structures for different energy ranges for a convergence criterion of 1% for the aforementioned reaction rates.

For the fast neutron dosimetry, we generated a 450-group LIB450 library that has only one group below 0.1 MeV. The LIB450 was collapsed into two broad-group structures, using the scalar flux [5] and bi-linear adjoint weighting [6] techniques. The 16-group, LIB16, and 15-group, LIB15, libraries were generated by the scalar flux and bi-linear adjoint weighting techniques, respectively. Table I(a) gives the broad- to fine-group reaction rate ratios for the six aforementioned dosimetry isotopes at the cavity dosimeter. Reaction rates calculated using the broad-group library are closely comparable (within ~1%) to those calculated using the fine-group library.

TABLE I. Broad- to Fine-Group Reaction Rate Ratios

<table>
<thead>
<tr>
<th>Response</th>
<th>LIB16/LIB450</th>
<th>LIB15/LIB450</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{63}\text{Cu}(n,\alpha)$</td>
<td>0.9882</td>
<td>1.006</td>
</tr>
<tr>
<td>$^{54}\text{Fe}(n,p)$</td>
<td>0.9884</td>
<td>0.9924</td>
</tr>
<tr>
<td>$^{58}\text{Ni}(n,p)$</td>
<td>0.9888</td>
<td>0.9946</td>
</tr>
<tr>
<td>$^{46}\text{Ti}(n,p)$</td>
<td>0.9896</td>
<td>1.005</td>
</tr>
<tr>
<td>$^{237}\text{Np}(n,f)$</td>
<td>0.9996</td>
<td>1.012</td>
</tr>
<tr>
<td>$^{238}\text{U}(n,f)$</td>
<td>0.9992</td>
<td>1.003</td>
</tr>
</tbody>
</table>

(a) For Fast Neutron Dosimetry

(b) For Thermal Neutron Gamma Production

<table>
<thead>
<tr>
<th>Response</th>
<th>LIB35/LIB589</th>
<th>LIB34/LIB589</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^1\text{H}(n,\gamma)^a$</td>
<td>0.9880</td>
<td>0.9984</td>
</tr>
<tr>
<td>$^{56}\text{Fe}(n,\gamma)^b$</td>
<td>0.9829</td>
<td>0.9920</td>
</tr>
</tbody>
</table>

a: calculated at mid-downcomer
b: calculated at 1/4 T pressure vessel

For thermal neutrons, we generated a 589-group, LIB589, library that has 449-fast, 83-epithermal, and 57-thermal groups. Two broad-group libraries were generated from LIB589: the 35-group, LIB35, by scalar flux weighting and the 34-group, LIB34, by the bi-linear adjoint weighting. Both broad-group libraries have 12 groups in the thermal energy range. Table I(b) presents broad- to fine-group reaction rate ratios for the gamma production from thermal neutrons considering $^1\text{H}$ and $^{56}\text{Fe}$. LIB35 and LIB34 libraries yield reaction rates which are in agreement (within ~1-2%) with those predicted by LIB589.

The last set of calculations consists of comparisons of broad-group to continuous energy reaction rates. The A\textsuperscript{3}MCNP (Automated Adjoint Accelerated MCNP) code [7] that is a version of MCNP with automated variance reduction capability, is used for continuous energy calculations.

Tables II(a) and II(b) give the broad-group to continuous energy reaction rate ratios, for the fast and the thermal energy range applications, respectively. For fast neutron dosimetry, besides the new libraries, we use Oak Ridge National Laboratory’s 47-neutron, 20-gamma group BUGLE-96 (B96) library [5]. For gamma production from thermal neutrons, the LIB35, LIB34 and BUGLE-96T (B96T) [5] libraries are utilized. The new broad-group libraries yield more accurate results than the BUGLE-96 library. For example, there is ~4% improvement in the $^{237}\text{Np}(n,f)$ reaction in fast neutron dosimetry calculations comparing LIB15 with BUGLE-96. For the $^1\text{H}(n,\gamma)$ reaction in the thermal energy range, LIB34 and LIB35 have less than ~6% difference, while BUGLE-96T shows a difference less than ~16% as compared to continuous energy predictions. It is worth noting that the new libraries have resulted in better prediction, while having almost half the number of groups used by the standard BUGLE-96 library.

TABLE II. Broad- to Continuous Energy Reaction Rate Ratios

<table>
<thead>
<tr>
<th>Response</th>
<th>LIB35/LIB589</th>
<th>LIB34/LIB589</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^1\text{H}(n,\gamma)^a$</td>
<td>0.9880</td>
<td>0.9984</td>
</tr>
<tr>
<td>$^{56}\text{Fe}(n,\gamma)^b$</td>
<td>0.9829</td>
<td>0.9920</td>
</tr>
</tbody>
</table>

CONCLUSION

We have developed the CPXSD methodology that constructs group structures for a problem and objective of interest. The CPXSD methodology was applied to a reactor pressure vessel problem using TMI-1 to generate new group structures for the fast neutron dosimetry applications, and for the thermal neutrons due to their production of gamma rays. It was demonstrated that the broad-group libraries containing the CPXSD generated group structures are in close agreement with their fine-group libraries (within 1-2%). Also, comparing with continuous energy Monte Carlo predictions, we have demonstrated that these new libraries yield more accurate results than the standard BUGLE libraries. For example, the $^{237}\text{Np}(n,f)$ fast neutron dosimetry reaction rate computed by the LIB15 library is ~4% closer to the
continuous energy results as compared to BUGLE-96. In the thermal energy range, for the $^{1}\text{H}(n,\gamma)$ reaction rate the maximum difference for new libraries is less than ~6%, while BUGLE-96 shows a maximum difference of less than ~16%. Our analyses indicate that the group structures constructed by the CPXSD methodology can significantly improve the efficiency and accuracy of shielding calculations.

TABLE II. Broad-Group to Continuous Energy (CE) Reaction Rate Ratios

(a) For Fast Neutron Dosimetry

<table>
<thead>
<tr>
<th>Response</th>
<th>LIB16 / CE</th>
<th>LIB15 / CE</th>
<th>B96 / CE</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{63}\text{Cu}(n,\alpha)$</td>
<td>.9301</td>
<td>.9056</td>
<td>.9111</td>
</tr>
<tr>
<td>$^{54}\text{Fe}(n,p)$</td>
<td>.9196</td>
<td>.9152</td>
<td>.9006</td>
</tr>
<tr>
<td>$^{58}\text{Ni}(n,p)$</td>
<td>.9167</td>
<td>.9168</td>
<td>.9000</td>
</tr>
<tr>
<td>$^{46}\text{Ti}(n,p)$</td>
<td>.9255</td>
<td>.9059</td>
<td>.9074</td>
</tr>
<tr>
<td>$^{237}\text{Np}(n,f)$</td>
<td>.9052</td>
<td>.9380</td>
<td>.8984</td>
</tr>
<tr>
<td>$^{238}\text{U}(n,f)$</td>
<td>.9063</td>
<td>.9219</td>
<td>.8990</td>
</tr>
</tbody>
</table>

(b) For Thermal Neutron Gamma Production

<table>
<thead>
<tr>
<th>Response</th>
<th>LIB35 / CE</th>
<th>LIB34 / CE</th>
<th>B96T / CE</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{1}\text{H}(n,\gamma)$&lt;sup&gt;ab&lt;/sup&gt;</td>
<td>.9449</td>
<td>.9516</td>
<td>1.157</td>
</tr>
<tr>
<td>$^{56}\text{Fe}(n,\gamma)$&lt;sup&gt;cd&lt;/sup&gt;</td>
<td>.9910</td>
<td>.9838</td>
<td>.9419</td>
</tr>
</tbody>
</table>

<sup>a</sup> A3MCNP statistical relative errors of fluxes are ≤ 1%
<sup>b</sup> calculated at back of downcomer
<sup>c</sup> A3MCNP statistical relative errors of fluxes are ≤ 2%
<sup>d</sup> calculated at ¼ T pressure vessel

REFERENCES