Strategic Assessment of Causes, Impacts & Mitigation of Radiation Embrittlement of RPV steel in LWRs

Jubair Ahmed Shamim [a], Palash Kumar Bhowmik [a], Abhinav Gairola [a], Kune Y. Suh [a, b*]
[a]*Department of Nuclear Engineering, Seoul National University, 1 Gwanak Ro, Gwanak Gu, Seoul 151-744, ROK*
[b]*PHILOSOPHIA Inc., 1 Gwanak Ro, Gwanak Gu, Seoul 151-744, ROK*
[*]*Corresponding author: kysuh@snu.ac.kr*

1. Introduction

Nuclear power has been emerged as a proven technology in the present day world to beget electricity after its first successful demonstration in 1942. Due to world's increasing concern over the augmented concentration of "Greenhouse Gas" emissions primarily caused by burning of fossil fuel, it is not surprising that there will be a galloping demand for nuclear power in near future.

As per data of World Nuclear Association, there are currently 435 operable civil nuclear power reactors around the world, with a further 71 under construction, among which the most common type is LWR. Pressure vessel of LWR is the most vital pressure boundary component of Nuclear Steam Supply System (NSSS) as it houses the core under elevated pressure and temperature. It also provides structural support to RPV internals and attempts to protect against possible rupture under all postulated transients that the NSSS may undergo.

LWR pressure vessel experiences service at a temperature of 250-320 ^{0}C and receives significant level of fast neutron fluence, ranging from about 5×10^{22} to 3×10^{24} n/m^2 depending on plant design. There are also differences in materials used for various designed reactors. Weldments also vary in type and impurity level. Accordingly, the assessment of degradation of major components such as RPV steel caused by aging and corrosion is a common objective for safe operation of all LWRs.

The purpose of this paper is to assess how neutron irradiation contributes to the degradation of mechanical properties of RPV steel and how these effects can be minimized. Since RPV is the only irreplaceable component in NPPs, the degradation of mechanical properties of RPV is the life-limiting feature of LWR nuclear power plant operation.

2. Aging Mechanisms of LWR Pressure Vessel Materials

RPV is the unique component of LWR nuclear power plants as it acts as the main barrier against radioactive leakage and limits the lifetime of NPPs. The major aging mechanisms of LWR pressure vessel materials can be categorized as follows:

(a) Radiation Embrittlement
(b) Thermal Aging
(c) Temperature Embrittlement
(d) Fatigue & Wear

In this study, the authors would like to concentrate on details of radiation embrittlement, how it degrades the mechanical properties of RPV steel and what are the mitigating ways to abate the effects of radiation embrittlement based on available literature.

3. Radiation Embrittlement

The degradation of mechanical properties of LWR pressure vessel steel adjacent to beltline region due to irradiation coming from the core is known as radiation embrittlement. The interaction of high energy subatomic particles and radiation with crystal lattices can give rise to a variety of defects, including vacancies and self-interstitials. The following figure delineates the process of radiation embrittlement at a glance:

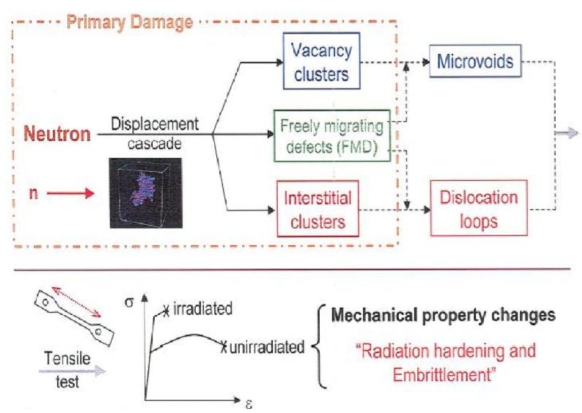

Fig. 1. Process of radiation embrittlement of LWR pressure vessel steel, after ref [1].

The microscopic causes of radiation embrittlement is mainly emerged from the forming of obstacles to dislocation motion in lattice crystal as well as change in composition and structure of microscopic interfacial regions along which crystal plane sliding occurs. Both these phenomena are due to radiation-matter interactions, among which most of damage is caused by fast neutrons in LWRs. γ-rays may also contribute to this causes but in less frequent circumstances. The mechanisms causing embrittlement is described in the subsequent sections of this paper.

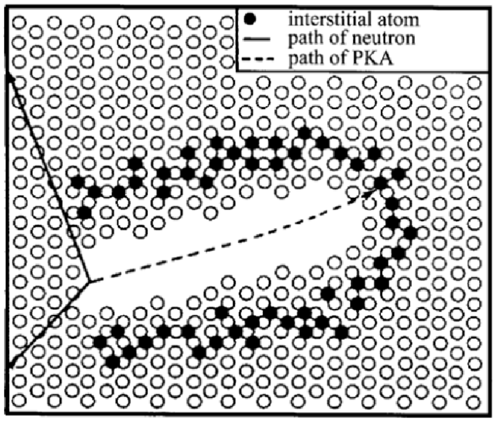

Fig. 2. Original version of the displacement spike. [After J. A. Brinkman, Amer. J. Phys., 24:251 (1956).]

3.1 Embrittlement by Neutron Irradiation

If an incident radiation (neutron) strikes an atom in crystal lattice with sufficient energy higher than it's the binding energy, the lattice atom will be displaced as a primary knock-on atom (PKA) leaving behind a vacant site (one vacancy and one PKA residing in lattice as interstitial constitutes a Frenkel pair). The PKA moves through lattice and creates further knock-on atoms in a displacement cascade which is illustrated in figure 2.

The number of displaced atoms can be approximated by Kinchin-Pease (K-P) model which is summarized in equation (1) and a graphical representation of the same is depicted in figure 3.

$$N_d = \begin{cases} 0 \text{ for } T < E_d \\ 1 \text{ for } E_d < T < 2E_d \\ \dfrac{T}{2E_d} \text{ for } 2E_d < T < 2E_c \\ \dfrac{E_c}{2E_d} \text{ for } T > E_c \end{cases} \quad (1)$$

where, N_d is the number of displaced atoms, T is the receipt of energy by struck atom, E_d is the energy required to displace the struck atom i.e. displacement threshold and E_c is the energy loss by electron stopping.

However, Mansur and Farrell [2] stated that thermal neutron induced displacements becomes significant ($\geq$10%) with respect to those from fast neutrons when:

$$\frac{\varphi_{nt}}{\varphi_{nf}} \geq 10 \quad (2)$$

where, φ_{nt} and φ_{nf} are the thermal and fast neutron flux respectively.

Neutrons can cause embrittlement of RPV also by introducing the ^{10}B (n, α) ^{7}Li reaction, which can be written as follows:

$$n + {}^{10}B \rightarrow {}^{4}He\ (1.775\ MeV) + {}^{7}Li\ (1.015\ MeV) \quad (3)$$

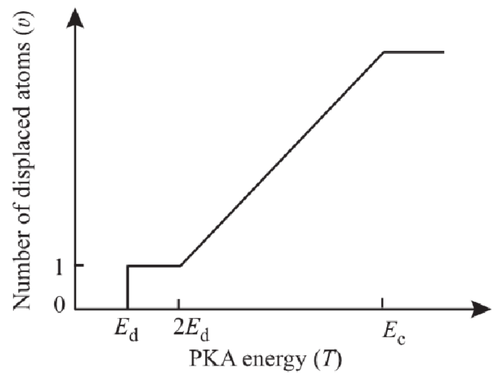

Fig. 3. The number of displaced atoms in the cascade as a function of the PKA energy according to the model of Kinchin and Pease, [taken from G. S. Was, Fundamentals of Radiation Materials Science, 2007].

Since boron is present in steel as an impurity and also sometimes included during manufacturing of various steels, neutron irradiation can yield helium production by reacting with boron. Thus the helium bubbles formed tends to coalesce at the grain boundaries, initiating the so called grain boundary cracking, which also contributes to embrittlement of RPV steel.

3.2 Embrittlement by Gamma-ray Irradiation:

Irradiation by γ-rays originating either from fissioning fuel in reactor core or from the interaction of core and peripheral structural materials with thermal and fast neutron may also contribute to displacement damage in RPV by transferring γ-energy to electrons via Compton scattering, pair production and photoelectric effect.

A graphical representation of above mentioned effects in in relatively light and heavy metals Al and Pb is shown in figure 4 where the result is expressed in terms of linear attenuation coefficient or macroscopic cross section.

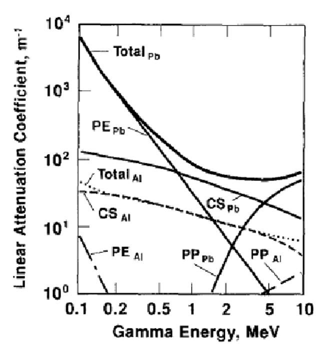

Fig. 4. Linear attenuation coefficient as a function of γ-ray energy for Al (Z=13) and Pb (Z=82), [taken from S. Glasstone, A. Sesonske, Nuclear Reactor Engineering, 1967].

Again, by using the cross section as comparison tool, Mansur and Farrell [2] concluded that γ-induced displacements contribute more than 10% compared to fast neutron induced displacements if the following condition is satisfied:

$$\frac{\varphi_{\gamma}}{\varphi_{nf}} \geq 100 \quad (4)$$

where, φ_{γ} is the γ-ray flux.

4. Effects of Irradiation on Mechanical Properties

The literature on irradiation effects in RPV is enormous and detailed presentation of those is beyond the scope of this paper. Hence, a brief overview of how neutron irradiation degrades mechanical properties of RPV steel has been described in the later paragraphs.

The low alloy RPV steels are ferritic steels that exhibit the classic ductile-to-brittle transition behavior with decreasing temperature. In the low toughness region, transgranular cleavage is the failure mode, while ductile rupture (shear fracture) is the failure mode in the high toughness region. As temperature increases from the low to high toughness region, the cleavage fracture probability decreases and the ductile rupture probability increases. Neutron irradiation tends to increase the temperature at which this transition occurs and tends to decrease the ductile toughness. The effect of neutron irradiation on mechanical properties of RPV steel is described through figure 5 to 7.

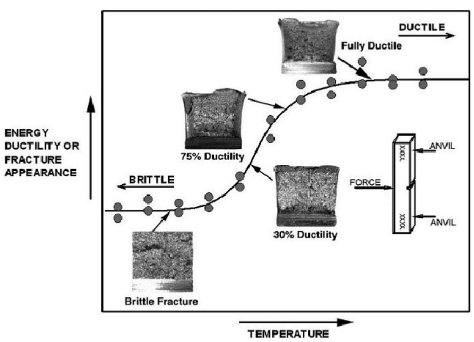

Fig. 5. RPV Steel Exhibit a Rapid DBT by Measuring the Energy to Break a CVN Specimen under Impact Loading, after ref [3].

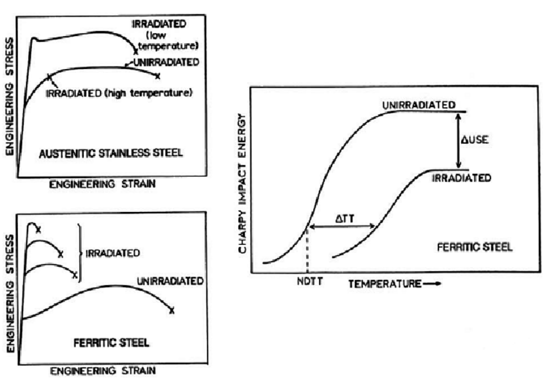

Fig. 6. Effects of increasing irradiation fluence on the tensile stress–strain curves for typical ferritic and austenitic stainless steels, as well as effects on Charpy impact energy, after ref [3].

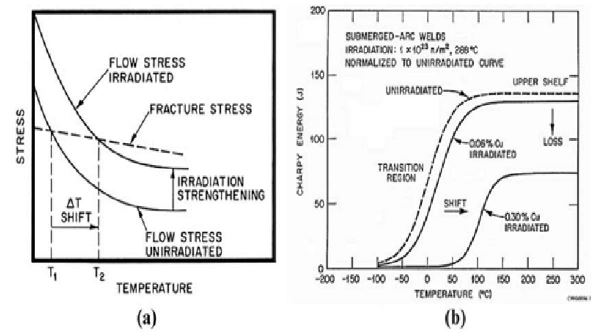

Fig. 7. Schematic diagrams depicting (a) how the irradiation-induced strength increase results in an upward shift in the Charpy impact toughness transition temperature and (b) showing the significant role of copper content towards increasing radiation sensitivity, after ref [3].

5. Quantitative Prediction of Embrittlement

As per recommendation of Section 50.61 of 10 CFR Part 50 [4], the following relation can be used to approximate ΔDBTT (^{0}C):

$$\Delta DBTT = \frac{5}{9}.\left[CF.f^{\,(0.28-0.10\log_{10} f)} + M \right] \quad (5)$$

where:

- f is the fast neutron fluence, in units of 10^{19} n/cm^{2}

- CF (ºF) is a "Chemistry Factor", dependent on Cu and Ni content. Because of the embrittlement-enhancement effect caused by these two elements, CF increases as the Cu and Ni content increases, ranging from 20ºF (0 wt-% Cu, 0 wt-% Ni) to 320ºF (0.4 wt-% Cu, 1.20 wt-% Ni)

- M (ºF) is a coefficient added to obtain conservative, upper bound values of the post-irradiation DBTT. It accounts for uncertainties in the analytical relations as well as in the knowledge of the pre-irradiation DBTT. A typical value for M is 56ºF for welds and 34ºF for base metal (after Shah, V.N., MacDonald, P.E.).

Equation (5) is not universally applicable, its use is recommended as long as [5]:

- the material under investigation is a ferritic steel of type SA-302, 336, 533, 508 (typical RPV steels);

- the minimum specified yield strength, in unirradiated conditions, is 345 MPa;

- the irradiation took place at a nominal temperature of 550ºF (288ºC).

6. Types of Defects Produced as a Result of Radiation Embrittlement

6.1 Black-Dot Structure:

The black spot defect cluster evolution in neutron-irradiated Cu at 70^{0}C is shown in figure 8. The defects produced at these conditions appear as black dots in the electron micrograph. As long as the irradiation temperature is below 350°C, increasing fluence simply increases the density of the black-dot damage. When irradiation is carried out at temperatures greater than 350°C, the nature of the microstructure is entirely different from the black-dot pattern characteristic of low-temperature irradiation.

6.2 Dislocation Loops:

Dislocation loops (figure 9) can form at lower temperatures ($T < 0.2\ T_m$, where T_m is the absolute melting temperature of the irradiated alloy). The displacement cascade can be thought of as a core of vacancies surrounded by a shell of interstitials. If the vacancy core or the interstitial shell collapse (condense) onto a close-packed plane, dislocation loops can be generated: collapse of the vacancy core results in a vacancy loop, whilst collapse of the interstitial shell results in an interstitial loop.

6.3 Voids:

Under some conditions the embryo collection of vacancies of can begin to grow in a 3D manner rather than collapse into a dislocation loop. This route leads to the formation of voids in metals and consequent swelling of the structure. Voids produced in Cu-100 ppm B irradiated at 182 ^{0}C (1 dpa) is shown in figure 10. The voids are not spherical. Rather, they assume the shape of a regular octahedron with {11 1} planes as surfaces. The ends of the octahedron, however, are truncated by {100} planes. Voids are annealed out of the microstructure at about 750°c.

6.4 Helium Bubbles:

At temperatures above 800 ^{0}C, dislocation loops and voids are not found in irradiated steel. In addition to grain boundaries, dislocations and carbide precipitates, the microstructure contains small helium-filled bubbles (figure 11). Helium is generated by (n, α) reactions with the boron impurity in the steel and with the major constituents, principally nickel. At temperatures below 650 ^{0}C, the helium atoms produced by stopping the alpha particles in the material are not mobile enough to migrate and nucleate bubbles.

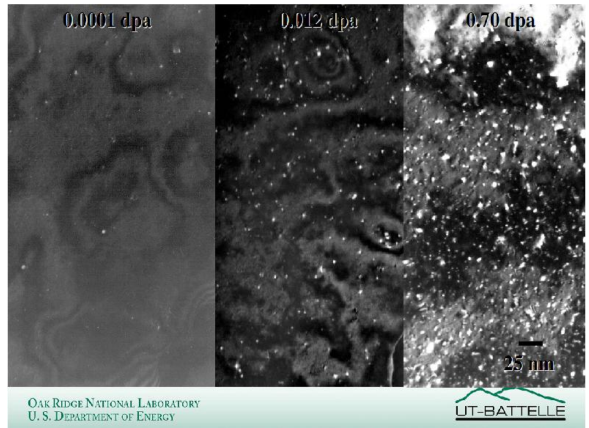

Fig. 8. Black Spot Defect Cluster in Neutron-Irradiated Cu at 70 ^{0}C, (after Steven J. Zinkle, Metals and Ceramic Devision, ORNL).

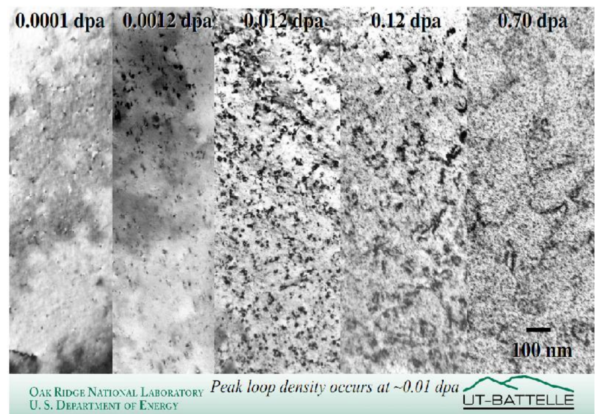

Fig. 9. Dislocation Loop in Neutron-Irradiated Cu at 70 ^{0}C, (after Steven J. Zinkle, Metals and Ceramic Devision, ORNL).

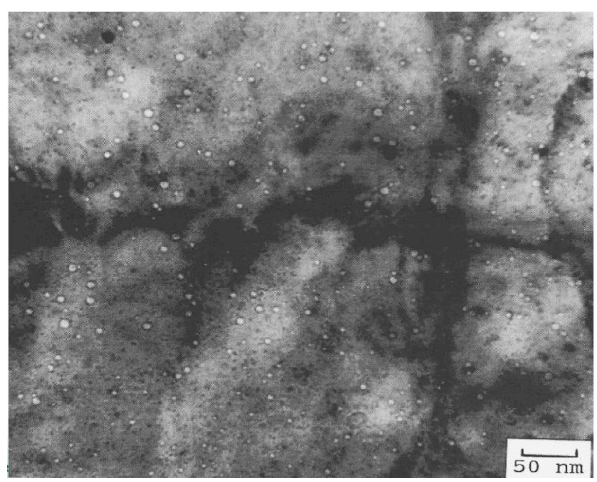

Fig. 10. Voids in Cu-100 ppm B irradiated at 182 ^{0}C (1 dpa), (after Steven J. Zinkle, Metals and Ceramic Devision, ORNL).

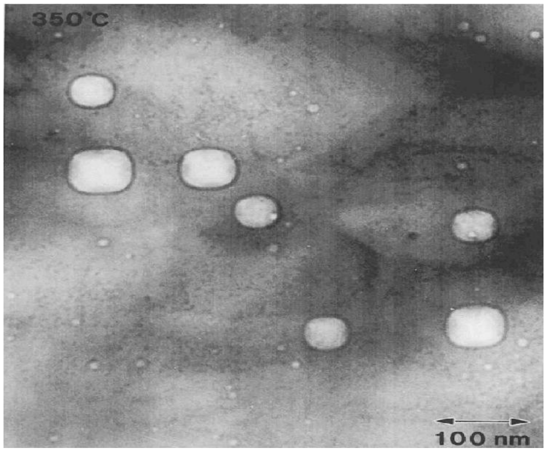

Fig. 11. Voids & He Bubbles in Cu-100 ppm B Irradiated at 350 ^{0}C in ORR Fission Reactor (1 dpa), (after Steven J. Zinkle, Metals and Ceramic Devision, ORNL).

7. Effects of Irradiation on RPV Operation

Integrity of RPV is maintained until the materials in the beltline region near the core of the reactor possesses adequate fracture toughness. Due to neutron irradiation adjacent to the beltline area, material toughness and tensile properties may undergo notable changes. As a result, structural integrity of RPV steel needs to be assessed periodically which is determined using three main elements:

(a) The material fracture toughness, which is a function of the operating environment;
(b) The mechanical and thermal stresses experienced during normal operating and severe accident transients;
(c) The size and potential growth of defects postulated (or measured) to be present in the RPV structure.

In order to assess integrity during normal operation, a reference defect is assumed to be present at either the inside or outside of the RPV wall, and the applied stresses are dependent on the operating transients involved. The most common normal operation transients are heat-up and cool-down, and the maximum operating pressure is calculated as a function of temperature using a lower bound fracture toughness curve (figure 12) indicative of the degree of embrittlement experienced by the RPV.

8. Mitigation of Radiation Embrittlement

The radiation embrittlement can be mitigated by either flux reductions (operational methods aimed at managing ageing mechanism) or by thermal annealing of the RPV (maintenance method aimed at managing ageing effects). Flux reductions can be achieved by either fuel management or shielding the RPV from neutron exposure.

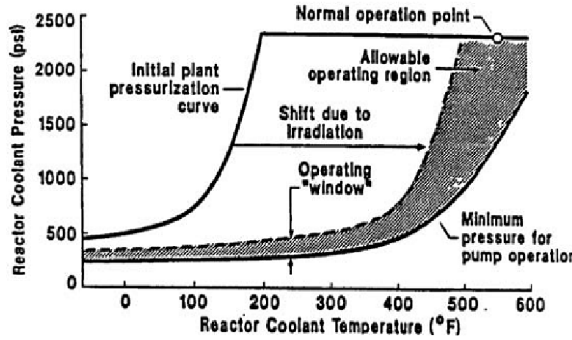

Fig. 12. Irradiation-Induced Shift in Operating P–T Curves and Operating Window, after ref [3].

8.1 Fuel Management:

The neutron flux (hence fluence) can be reduced by initiating a fuel management program early in the life of a given plant. Such fuel management is carried out by implementing a low neutron leakage core (LLC). A LLC is a core that utilizes either spent fuel elements or dummy (stainless steel) fuel elements on the periphery of the core which reflect neutrons back into the core or absorb them rather than allowing them to bombard the RPV wall. LLCs can result in a reduction in power and/or increase in cost to the NPP owner.

A more drastic reduction of neutron flux can be achieved by inserting shielding dummy elements into the periphery of an active core. Dummy elements were inserted into most of the WWER- 440/V-230 reactors in the middle of the 1980s. Thirty-two dummy elements are usually inserted into the core periphery. They cause not only a significant flux reduction but also a shifting of the maximum neutron flux by an angle of about 15° relative to both sides of the hexagon corners. Thus 12 new peak values of neutron flux are created on the pressure vessel wall. The original peak flux is decreased by a factor of 4.5 and the "new" peak flux is decreased by a factor of close to 2.5 shown in figure 13. Thus, the cumulative effect of flux reduction must be calculated for both locations. Again, this method is most effective when applied during the first years of operation or just after a thermal annealing. The use of dummy elements usually results in a significantly different neutron balance in the core. The radial gradient is increased and thus the power distribution is disturbed in such a way that the peak power may exceed certain limits. Thus, a reduction in the fuel cycle length or a reduction of the reactor output are often necessary.

8.2 RPV Shielding:

The intensity of flux (hence fluence) can also be lessened by providing additional shielding to the RPV wall which protects it against neutron bombardment. Shielding of the RPV wall can be done by increasing the thickness of the thermal pads that exist on the thermal shield at locations where the fluence is high or by placing shielding on the RPV wall. A number of alloys or elements are available to provide shielding of the RPV wall by absorbing the high energy neutrons among which tungsten is considered as most effective.

8.3 Thermal Annealing:

Once a RPV is degraded by radiation embrittlement (e.g. significant increase in Charpy ductile-brittle transition temperature or reduction of fracture toughness), the only way to recover the RPV material toughness properties is thermal annealing. In this method the RPV (with all internals removed) is heated up to some temperature with the help of an external heat source (e.g. electrical heaters, hot air), held for a certain period and then slowly cooled down.

In the early 1980s, the Westinghouse Electric Corporation performed a study to examine the feasibility of thermal annealing of commercial RPVs and developed an optional, in situ, thermal annealing methodology that maximizes the fracture toughness recovery, minimizes re-exposure sensitivity and minimizes reactor downtime when thermal annealing becomes essential. It was concluded from this study that excellent recovery of all properties could be achieved by annealing at a temperature of some 450°C (850°F) or higher for 168 hours. Such an annealing was predicted to result in a significant ductile-brittle transition temperature recovery.

Revisions to 10 CFR 50.61 and 10 CFR 50 Appendices G and H, new section 10 CFR 50.66 (the thermal annealing rule) and new Regulatory Guide 1.162 has been issued by the USNRC which explicitly cites thermal annealing as a method for mitigating the effects of neutron irradiation, thereby reducing RT_{PTS}.

9. Summary and Conclusions

Although there are a number of ways (e.g. thermal neutrons, fast neutrons and gamma-ray irradiation) that may contribute to the displacement of atoms (hence RPV embrittlement and degradation of mechanical properties), most of the literatures relates the severity of embrittlement to fast neutron fluence only, neglecting the irradiation time and effect of gamma-rays. Irradiation rate becomes significant when RPV walls experience a low fast neutron flux and gamma-rays plays a prominent role when a thick water gap separates the core from RPV walls. Therefore, in order to ensure adequate safety and proper life time of RPV, all possible sources of atom displacement should be taken into consideration for each material of interest in the particular irradiation environment.

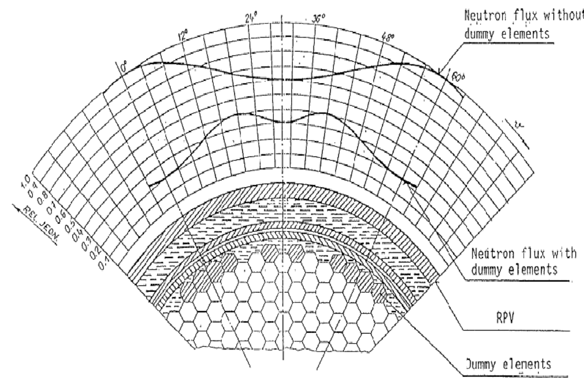

Fig. 13. WWER (Water, Water, Energy Reactor) Flux distributions in low leakage cores (taken from IAEA-TECDOC-1120)

ACKNOWLEDGEMENTS

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP) (No. 2008-0061900) and partly supported by the Brain Korea 21 Plus Project (No. 21A20130012821).

REFERENCES

[1] H. Hannu, Material Development in New Reactor Designs- Gen III and SCWR Concept, 20th International Conference on Structural Mechanics in Reactor Technology (SMiRT) August 9 - 14, 2009, Dipoli Congress Centre, Espoo, Finland.
[2] L.K. Mansur, K. Farrell, Mechanisms of Radiation-Induced Degradation of Reactor Vessel Materials, Journal of Nuclear Materials, vol. 244, pp. 212-218, 1997.
[3] "Integrity of Reactor Pressure Vessels in Nuclear Power Plants: Assessment of Irradiation Embrittlement Effects in Reactor Pressure Vessel Steels", IAEA Nuclear Energy Series, No. NP-T-3.11, pp 24-43, 2009.
[4] Code of Federal Regulations, 10 CFR Part 50 "Domestic Licensing of Production and Utilization Facilities", US-NRC. Last Revised in August 2006.
[5] Regulatory Guide 1.99, "Radiation Embrittlement of Reactor Vessel Materials", Rev. 2, May 1988. US-NRC.