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Some recent research results and their implications for RPV irradiation surveillance under long term operation

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ABSTRACT

During the last decade the irradiation behaviour of characteristic RPV steels used in Pressurized Water Reactors has been extensively investigated in the international research programs CARISMA, CARINA and LONGLIFE. Whereas the CARISMA and CARINA programs were mainly focused on material testing of German RPV steels and data base applications by the RT_{NDT} and Master Curve concepts, the main emphasis of the LONGLIFE project was placed on microstructural examinations of a wide range of RPV steels used in European NPP. Atom Probe Tomography, Small Angle Neutron Scattering, Transmission Electron Microscopy and others belonged to the techniques applied.

Four of those RPV steels which are particularly interesting under the point of view of long term irradiation behavior have been studied at different irradiation levels up to 8×10^{19} n/cm^2 (E > 1 MeV) in all three research programs. In this way it was possible to get a comprehensive understanding of the material behavior induced by neutron irradiation and the responsible mechanisms behind. By means of the material properties and microstructural data obtained, in particular the impact of chemical composition (copper and nickel), and neutron flux on the material's fracture toughness was quantified. Moreover, the most important mechanisms such as formation of element specific precipitations, segregations, and matrix defects could be demonstrated in relation to the change of material properties. For one material the occurrence of late irradiation effects has been observed in both material properties and microstructural appearance.

All the available data are discussed thoroughly by consulting the relevant literature in the light of the practical implications they have for RPV irradiation surveillance under long term operation of 60 years and beyond. Specific issues like material specific application of reference temperature concepts, data scattering, prediction of high fluence behavior and use of material test reactor data are addressed. Finally, based on the results obtained by the research programs concerned, best practices for RPV irradiation surveillance programs are suggested and exemplified by industrial experience and adopted measures already in use.

KEY WORDS

Pressurized Water Reactor, RPV integrity, long term operation, irradiation surveillance program, fracture toughness, irradiation embrittlement, Master Curve, RT_{NDT} concept

INTRODUCTION

The proof of a sufficient safety margin against brittle fracture of the reactor pressure vessel (RPV) is essential for the operational safety of nuclear power plants because the RPV is a key component with respect to the management of deviations from normal operation, the maintenance of core cooling and the general function for fission product retention. The common procedure to be applied to prove the RPV safety involves a concept developed in the USA in the 1970s – the RT_{NDT} concept – which is based on the definition of a deterministic lower bound fracture toughness – temperature curve obtained from measured data. This curve is then adjusted to the material concerned with the aid of the adjusted reference temperature RT_{NDTj} with T_{41} shift from Charpy tests taking into account the neutron irradiation. The position of the adjusted fracture toughness curve must be determined either indirectly according to the RT_{NDT} concept or directly according to the fracture mechanics concept by testing irradiated fracture mechanics specimens with subsequent determination of an appropriate reference temperature based on these test results.

This fracture mechanics concept, which is based on the so-called Master Curve or T_0 concept [1], is being further advanced and applied worldwide as an appropriate method for the RPV safety assessment against brittle failure. The analysis of the measured fracture toughness data is performed by a statistical approach that allows a probabilistic evaluation of the curve created with this method. The 50 % fractile of the fracture toughness curve normalized to 1T (25 mm) specimens – the Master Curve – is used to determine a brittle fracture transition temperature T_0 at 100 $MPa{\cdot}m^{0.5}$, which may be used with a margin as reference temperature $RT_{T0} = T_0 + 19.4$ K for the adjustment of the lower bound fracture toughness – temperature curve [2]. An essential advantage of the Master Curve concept is the direct determination of the reference temperature for brittle fracture by fracture mechanical tests and therefore a more realistic transfer to the component behavior. In this context there is a strong interest to further validate the Master Curve based RT_{T0} procedure by appropriate fracture toughness data of representative irradiated RPV materials.

Whichever reference temperature (RT_{NDTj} or RT_{T0}) is applied the reference temperatures of the monitored RPV materials have to be determined by means of a RPV irradiation surveillance program or an equivalent program. Usually the reference temperature, which is a parameter for the change of material properties in terms of fracture toughness, increases with increasing fast neutron irradiation (fluence), however in particular in the long term operation (LTO) mode the measured data shows an increasing deviation from the predictions, as a consequence of high scatter or any other unexpected deviation. In particular the impact of copper and nickel, and neutron flux on the material's fracture toughness and possible late irradiation effects are of interest. In order to clarify these issues a sound understanding of the mechanisms behind is required. In this context microstructural examinations of tested irradiated RPV materials provide insight into the irradiation induced changes of the material matrix and the occurrence of so-called nanofeatures down to the atomistic scale. In order to correlate the material properties with the microstructural data well characterized materials at several fluence levels have to be studied, and a sufficiently broad and representative data base is required.

For this reason a number of irradiated and unirradiated original RPV materials used in Pressurized Water Reactors (PWR) designed by former Siemens/KWU company have been investigated in the CARISMA [3] and CARINA programs [4] which were mainly focused on material testing. Four of those RPV steels, one base material and three weld metals, which are particularly interesting under the point of view of long term irradiation behavior have been studied at fluences up to 8×10^{19} n/cm^2 ($E > 1$ MeV) in the European research project LONGLIFE by various microstructural techniques such as Atom Probe Tomography (APT), Small Angle Neutron Scattering (SANS), Transmission Electron Microscopy (TEM), Positron Annihilation Spectroscopy (PAS), and Auger Electron Microscopy (AES).

The irradiation induced change of material properties and the microstructural data obtained for the four RPV steels concerned are discussed with respect to the main impact parameters regarding the irradiation behavior and the most important mechanisms behind, with the aim to suggest best practices for RPV irradiation surveillance programs.

THE RESEARCH PROGRAMS CARISMA, CARINA AND LONGLIFE

CARISMA

The research program CARISMA (Crack Initiation and Arrest of Irradiated Steel Materials) which was finished in 2008 has produced a large data base to characterize the fracture toughness of pre-irradiated original RPV materials for a neutron fluence range of 4×10^{18} to 5×10^{19} cm^{-2} (E > 1 MeV) which is near or beyond the designed EoL range of German PWR. Three forged base materials (BM) and four weld metals (WM) as shown in Table 1 were subject of comprehensive tensile, Charpy-V impact, crack initiation and crack arrest tests in this program. The RPV materials concerned are representative for all four German PWR construction lines built by former Siemens/KWU company:

- Stade (KKS), Obrigheim (KWO) – 1st construction line (decommissioned 2003, 2005)
- Biblis A (KWB-A), Biblis B (KWB-B), Unterweser (KKU), Neckarwestheim 1 (GKN1) – 2nd construction line (decommissioned 2011)
- Grafenrheinfeld (KKG), Grohnde (KWG), Phillipsburg 2 (KKP2), Brokdorf (KBR) – 3rd construction line
- Emsland (KKE), Isar 2 (KKI2), Neckarwestheim 2 (GKN2) – 4th construction line

Table 1: Overview on CARISMA materials

Material	German PWR Generation	Material Code	Cu [wt%]	P [wt%]	Ni [wt%]
20MnMoNi5-5 JSW	4 (Konvoi)	P141 BM	0.05	0.01	0.79
22NiMoCr3-7 Klöckner	1-2	P7 BM	0.12	0.02	0.97
22NiMoCr3-7 JSW	3-4	P147 BM	0.05	0.01	0.84
S3NiMo1/OP 41 TT UP, GHH	4 (Konvoi)	P141 WM	0.03	0.02	1.01
S3NiMo3/OP 41 TT UP, Uddcomb	3	P16 WM	0.08	0.012	1.69
NiCrMo1 UP/LW320, GHH	2-4	KS05 WM	0.05	0.01	0.91
NiCrMo1,UP (modified)/ LW320, LW330	1	P370 WM	0.22	0.02	1.11

Most of the studied RPV steels were of optimized chemical composition, however one weld with high copper content (P370 WM), and one with high nickel content (P16 WM) respectively, were also examined. The CARISMA materials except the material KS05 WM were irradiated in a German test reactor, the VAK (Versuchsatomkraftwerk Kahl), in the 1980s in the frame of a dedicated irradiation program in order to make provisions for future changes of RPV safety requirements and possible Long Term Operation (LTO) measures. The irradiation temperature was mainly in a range between 280 °C and 290 °C. The RT_{NDT} concept and the Master Curve approach were applied for the assessment of the generated data in order to compare both approaches. A further objective was to clarify in which extent crack arrest curves can be generated for irradiated materials and how crack arrest can be integrated into the Master Curve approach. More detailed information on irradiation facilities, scope of testing and test results of the CARISMA program can be found in [3].

CARINA

The successor program CARINA (Extension of the Data Base of Fracture Mechanical Characteristics of Irradiated German RPV Materials) [4] was launched in 2008 and finished in 2012. The main objective of the CARINA program was to extend the experimental database generated by CARISMA to reach a final comparative assessment of the RT_{NDT} and Master Curve concepts for the proof against RPV brittle fracture for further irradiated German RPV materials including a larger fluence range together with special influences such as neutron flux, manufacture effects and specific irradiation effects, e.g. late blooming. The analysis of materials irradiated at higher neutron fluences did complement the representativity of the conclusions from previous studies for the applicability to the Nuclear Power Plants (NPPs) in Germany and some neighboring countries in terms of “upper bound” coverage for a fluence range covering more than the designed lifetime of the NPPs concerned. For all PWRs operated in 2013 in Germany the fluence for 32 EFPY is expected lower than 5×10^{18} (E > 1 MeV). In this way CARINA made a significant contribution in proving the safety of particularly German nuclear power plants and maintained the link to international procedures, especially with regard to the material behavior of reactor pressure vessels under neutron irradiation for long-term operation.

Three base materials (forgings), two heat affected zones (HAZ) and three weld metals have been studied as shown in Table 2 where the chemical composition of the specimen materials in terms of the most important chemical elements is also given. The RPV materials of the CARINA test matrix are representative for all four German PWR construction lines built by former Siemens/KWU company and due to the similarities of the vessel steels also for the German BWR type 72 at which the RPV is of significantly lower neutron flux.

Table 2: Overview on CARINA materials

Material	**German PWR Generation**	**Material Code**	**Cu [wt%]**	**P [wt%]**	**Ni [wt%]**
20MnMoNi5-5 JSW	4 (Konvoi)	P142 BM	0.06	0.005	0.8
22NiMoCr3-7 JSW	3	P150 BM	0.05	0.008	0.83
22NiMoCr3-7 JSW	3	P150 HAZ	-	-	-
22NiMoCr3-7 Klöckner	1-2	P151 BM	0.09	0.006	0.96
22NiMoCr3-7 Klöckner	1-2	P151 HAZ	-	-	-
Molytherme Electrode Sulzer	2	P152 WM	0.02	0.014	0.09
S3NiMo1/OP41TT GHH	4 (Konvoi)	P142 WM	0.06	0.012	0.9
S3NiMo3/OP41TT Uddcomb	3	P16 WM	0.08	0.012	1.69

The CARINA materials were irradiated in two facilities, in the VAK reactor and in a commercial PWR of Siemens/KWU design by means of an RPV irradiation surveillance program. The surveillance specimens were irradiated in standard capsules inserted in the RPV at a position of highest flux with a very low axial flux gradient near core midplane, and in so called gradient capsules at a position with lower flux due to the axial flux gradient near core edge.

LONGLIFE

LONGLIFE (Treatment of long term irradiation embrittlement effects in RPV safety assessment) is a 4 years collaborative research project launched in 2010 in the EURATOM part of the 7th Framework Programme of the European Commission [5]. The consortium consists of 16 European research and industrial companies. The project aims at improved knowledge on LTO phenomena relevant for European LWRs, the assessment and proposed improvements of prediction tools, codes and standards, and the elaboration of best practice guidelines on RPV irradiation surveillance. For this purpose the following six phenomena are studied by means of preselected RPV materials and congeneric model alloys:

- The high fluence behavior in terms of any additional damage mechanisms, which were not yet observed at fluences corresponding to the designed EOL of existing reactors (i.e. up to 40 years of operation). The high fluence behavior is also related to the appropriateness of existing trend curves for irradiation-induced changes in material properties at fluences corresponding to an extended EOL of 80 years.
- The neutron flux effect as an important issue in the perspective of using irradiation data from material test reactors and surveillance irradiation data from power reactors to predict the hardening and embrittlement behavior of RPV materials in the core belt region.
- The so-called late blooming effect (LBE) which may occur at high fluences, preferably in low Cu steels which contain significant amounts of Ni and Mn. These late irradiation effects might have an implication for RPV safety assessment under LTO conditions.
- The effect of alloying and impurity elements on the irradiation-induced changes in material properties such as Ni, Mn, Cu and Si.
- The thermal ageing of RPV steels as a general degradation mechanism connected with long term operation at high temperature may have some synergism with radiation effects for some steels.
- The issue of P-segregation and non hardening embrittlement may be important for RPV steels with higher P-content, however is not yet clear.

The experimental investigations in terms of material testing and microstructural analyses are based on 16 pre-selected materials classed into various RPV material groups with 7 base materials and 9 weld metals with significant sensitivity for specific long term irradiation effects, see Table 3. Most of the pre-selected materials are representative of Western and Russian LWRs in operation.

Among these materials there are four materials which have been used before in the CARISMA/CARINA investigations. Within LONGLIFE those materials have been investigated by APT, SANS, TEM, PAS and AES:

- P141 WM labeled ANP-2 in LONGLIFE
- P147 BM labeled ANP-4 in LONGLIFE
- P370 WM labeled ANP-5 in LONGLIFE
- P16 WM labeled ANP-6 in LONGLIFE

Some weld materials with higher contents of irradiation embrittlement promoting chemical elements are also considered (~0.2 % Cu in VTT-1 and ANP-5; ~1.6-1.7 % Ni in ANP-6, RAB-1 and NRI-1).

Table 3: Overview on LONGLIFE materials [5]

Code	**Type**	**Description**	**Irradiation condition, comment**	**T_{irr} (°C)**	**Max. fluence E >1 MeV (cm^{-2})**	**Testing techniques**
ANP-2	WM	S3NiMo1	MTR	285	5×10^{19}	AP, SANS, TEM, FT
ANP-6 RAB-1	WM	S3NiMo	MTR/Surveillance	280…300	6.8×10^{19}	AP, SANS, TEM, PAS
ANP-5	WM	NiCrMo1	MTR, evidence for flux effect	285	2.2×10^{19}	SANS, PAS
ANP-4	BM	22NiMoCr3-7	MTR	285	4.2×10^{19}	SANS, TEM
SCK-6	BM	22NiMoCr3-7	MTR (BR2)	290	16×10^{19}	SANS, TEM, Tensile
EDF-2	BM	16MND5	Surveillance	286	6.9×10^{19}	AP, SANS, TEM
EDF-3	WM	16MND5	Surveillance	286	5.1×10^{19}	AP, SANS, TEM
FZD-1a	BM	A533B high P (JPB)	MTR, evidence for LBE MTR (BR2)	255 290	9×10^{19} 15×10^{19}	AP, SANS, TEM, AES Tensile
FZD-1b	BM	A533B low P (JPC)	MTR, evidence for LBE MTR (BR2)	255 290	9×10^{19} 12×10^{19}	AP, SANS, TEM, AES Tensile
FZD-2	WM	10KhMFT	Decomm., Trepan Greifswald 4	265	3.1×10^{19}	SANS, AES FT, SPT, Tensile, HV
VTT-1	WM	10KhMFT	Surveillance	270	18×10^{19}	SANS, TEM FT
AEK-1	BM	15Kh2MFA	Surv. + MTR	270…290	145×10^{19}	AP, SANS, TEM, FT, HV
NRI-6	BM+ WM	15Kh2MFA	Surveillance	270	30×10^{19}	TEM
NRI-1	WM	5Kh2NMFAA	MTR	290	10×10^{19}	AP, TEM, Impact, Tensile, HV

In general the material behavior of the materials summarized in Table 1 and Table 2 is well known in terms of macrophysical material properties. However, the knowledge of the irradiation-induced changes in terms of microstructure is still poor except for the high copper material P370 WM where some Small Angle Neutron Scattering (SANS) results are available [6]. The availability of such microstructural data in particular for LTO fluences is essential for the understanding of the involved mechanisms and to improve the RPV irradiation surveillance concepts and prediction formulas.

RPV STEELS STUDIED

One base material P147 BM and three weld metals P141 WM, P370 WM and P16 WM of different chemical composition have been provided for microstructural examination in LONGLIFE (see Table 4). The weld metals are among those used in the four different constructions lines of German PWRs and represent high copper, high nickel and low copper/nickel welds, all manufactured by submerged arc welding (SAW). The low copper/nickel/phosphorus base material P147 BM is relevant to the newer PWR construction lines which are still in operation in Germany. All four materials were extensively tested within the research programs CARISMA and CARINA where a comprehensive database of mechanical properties was generated by a wide range of tensile, Charpy, drop-weight, fracture toughness and crack arrest tests. Moreover, there are some earlier SANS measurement results for the P141 WM and P370 WM material available [6]. Apart from the 0.22 % Cu P370 WM material which was manufactured by Klöckner Werke AG company for a research program to study different copper contents, all weld metals were manufactured from original wires and powders used for RPV irradiation surveillance samples. The P147 BM material was taken from a forged shell ring manufactured from Japan Steel Works Ltd. for a 900 MW class NPP with an inner diameter of 4.6 m and a wall thickness of approximately 230 mm. The 1.69 % Ni weld P16 WM was manufactured by former Uddcomb company. All four materials were irradiated in the 1980s in the German experimental Nuclear Power Plant in Kahl (VAK) at an irradiation temperature of roughly 285 °C. The neutron spectrum of the VAK reactor was comparable to that of other PWRs manufactured by Siemens/KWU; the neutron flux density at the capsule positions was approximately 2×10^{12} n/cm^2/s (E >1 MeV). In addition, some samples of P370 WM material were irradiated in a commercial PWR.

Table 4: Materials studied in both CARISMA/CARINA and LONGLIFE

Material	Type	Wire/powder base material	Chemical Composition [wt.%]									
			C	Si	Mn	P	S	Cr	Mo	Ni	Cu	V
P141 WM	Weld metal	S3NiMo1/ OP41TT	0.05	0.12	1.08	0.019	0.009	<0.10	0.62	1.01	0.03	0.01
P147 BM	Forging	22NiMoCr3-7	0.21	0.22	0.85	0.006	0.006	0.39	0.55	0.84	0.05	<0.01
P370 WM	Weld metal	NiCrMo1/ LW320/330	0.08	0.15	1.14	0.015	0.013	0.74	0.6	1.11	0.22	0.01
P16 WM	Weld metal	S3NiMo/ OP41TT/A 2	0.05	0.15	1.41	0.012	0.007	0.07	0.46	1.69	0.08	0.004

DISCUSSION OF RESULTS

High Cu steels and flux effect

For the high Cu weld P370 WM the T_{41} shifts at the same fluence but for two different fluxes are shown in Figure 1, together with high flux data of other high Cu welds.

No saturation effect at lower fluences is observed with respect to the irradiation embrittlement in terms of ΔT_{41}, however after a strong increase of ΔT_{41} at lower fluence a possible saturation plateau at about 160 K at higher fluences is observed for the P390 WM material. Taking into account the rather high Ni content (~1 %) of the high Cu welds concerned, for high Cu welds with low Ni content (~0.1 %) a saturation effect at lower fluences would be expected since the Cu precipitates rather fast and almost no Ni is available for further cluster formation.

Another point of interest is the potential impact of neutron flux on the irradiation behavior. The P370 WM results do not show any relevant flux effect on ΔT_{41} as can be seen in Figure 1.

As far as the CARINA/CARISMA materials and other German RPV steels are concerned no flux effect was observed with respect to the material properties [7]. However for other RPV steels irradiated at lower temperature of 270 °C a significant flux effect in Charpy transition temperature shift was observed for RPV materials with > 0.13 % copper content [8].

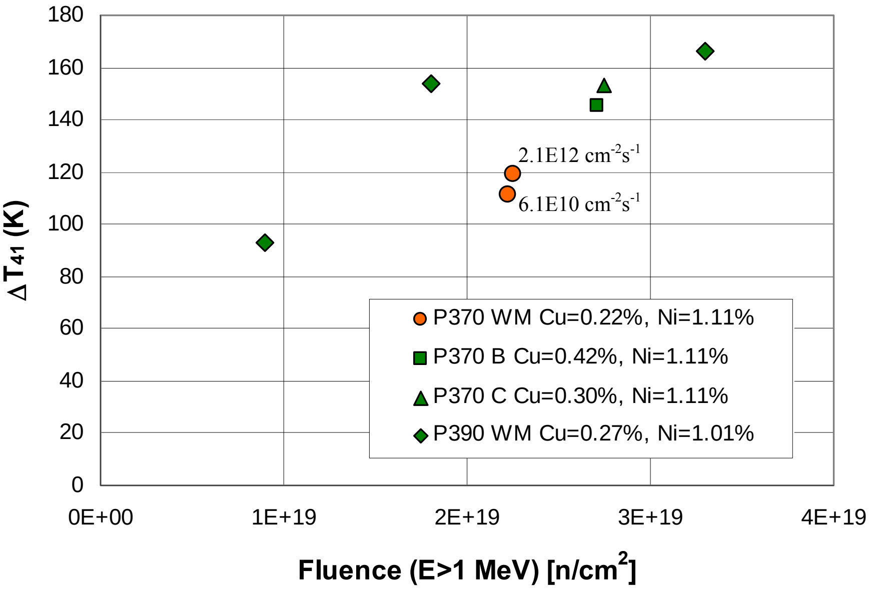

Figure 1: Measured ΔT_{41} for high Cu weld metals

The high copper P370 WM material was analyzed by SANS measurements performed by HZDR Dresden with the aim to clarify the issue if the flux independent behavior of material properties can be confirmed by microstructural insights or not.

As summarized in Table 5 a significantly strong effect of neutron flux on cluster size and number density was observed by SANS results obtained. The SANS results of the P370 MW material irradiated at two different levels of neutron flux (differing by a factor of 35) up to the same fluence shows that more cluster growth (by a factor of 2) occurred for the irradiation at the low flux [6]. However, the volume fraction of irradiation-induced clusters is almost the same and this gives an explanation that the material properties differ not much. This confirms also the general correlation of the irradiation-induced shift of the brittle–ductile transition temperature and the square-root of the volume fraction of irradiation-induced clusters, e.g. [9].

Table 5: Irradiation-induced clusters for P370 WM measured by SANS [6] and T_{41} data [3]

Material	Fluence, Φ (10^{19} cm^{-2}) (E>1MeV)	Flux, □ (10^{11} $cm^{-2}s^{-1}$ (E>1MeV)	Volume fraction, c (vol%)	Number density, N (10^{16} cm^{-3})	Radius, R_{mean} (nm)	Radius, R_{peak} (nm)	T_{41} (°C)
P370 WM	2.2	21	0.51±0.02	170±30	0.81	0.85	107
	2.2	0.6	0.53±0.02	50±6	1.16	1.60	99

Thus, as far as the high Cu material P370 WM is concerned, no significant flux effect on material properties was found, but a flux effect on microstructure in terms of number density and size of irradiation-induced clusters was observed.

This is partly in contradiction to other published data e.g. in [11] where RPV steels were irradiated in a material test reactor (MTR), and changes in Charpy transition temperatures as well as microstructural characteristics in terms of solute atom clustering were studied. In [11] was reported that high Cu material irradiated in a MTR showed larger shifts than those of surveillance data, while low Cu materials showed similar ones. However, the microstructural changes caused by MTR irradiation and surveillance irradiation were different, but it was pointed out that among others additional information on the comparison between MTR data and surveillance data are necessary for quantitative evaluation of flux effect. For example any spectrum effect was not addressed.

Late irradiation effects

For the material with the lowest Cu (0.03 %) and medium Mn/Ni (1.08/1.01 %) contents P141 WM both the Charpy T_{41} and the fracture mechanics T_0 data indicate an unexpected accelerated irradiation reaction at higher fluence as shown in Figure 2. Since such a behavior can be hardly explained by data scattering only and similar results have been observed even in a few RPV surveillance data [12], a confirmation of this observation by microstructural data was of interest.

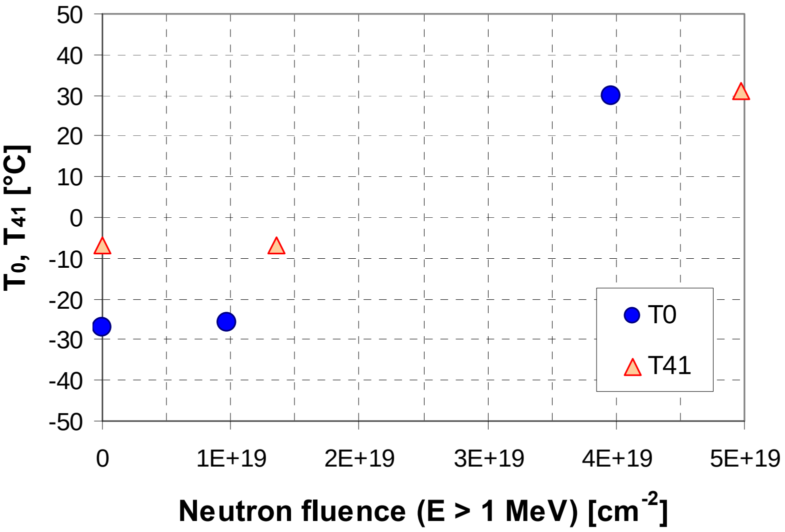

Figure 2: Measured T_{41} and T_0 data for P141 WM

Therefore, some preliminary microstructural data, which have been made available for the P141 WM material in the LONGLIFE project [10], is reviewed in the following. First, the development of Mn/Ni/Si/Cu clusters with fluence was analyzed by APT performed by Rolls-Royce. In the unirradiated material three precipitates containing these elements were found but this corresponds to an extremely low number density ($<3x10^{21}m^{-3}$) and a volume fraction of ~0.005% only. Even at $\sim1x10^{19}n/cm^2$ no evidence was found for the development of a significant number of Mn/Ni/Si/Cu clusters following the irradiation. However, at by the highest fluence of $\sim5x10^{19}n/cm^2$ a significant number of these features had formed (see Figure 3) with an estimated number density of $\sim3.6x10^{23}m^{-3}$ and a volume fraction of ~0.1% [13].

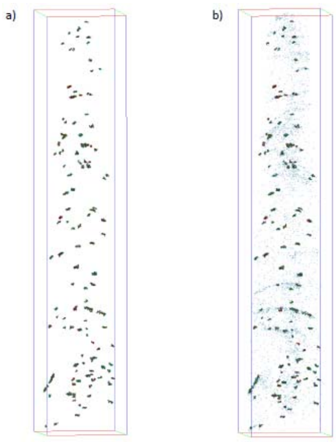

Figure 3: Mn/Ni/Si/Cu enriched clusters in P141 WM irradiated to $\sim5x10^{19}$ n/cm^2 (a) only clusters, (b) P distribution also shown [10]

The results from the SANS measurements performed by HZDR Dresden are shown in Table 6. Again, the fluence depending change in material properties (T_{41}, T_0) can be confirmed by the observed strong increase of number density and volume fraction.

Table 6: Irradiation-induced clusters for P141 WM measured by SANS [10]

Material	Fluence, Φ (10^{19} cm^{-2}) (E>1MeV)	Volume fraction, c (vol%)	Number density, N (10^{16} cm^{-3})	Radius, R_{mean} (nm)	Radius, R_{peak} (nm)	A-ratio
P141 WM	1.36	<0.005	2±2	-	-	~1
	4.70	0.10±0.02	55±5	0.72	0.55	2.54±0.08

Based on the increase of number density and volume fraction measured by both APT and SANS the fluence depending change in material properties (T_{41}, T_0) was confirmed.

<u>High nickel effect</u>

For the material with high Ni content P16 WM (1.69 % Ni, 1.41 % Mn, 0.07 % Cr) both the Charpy T_{41} and the fracture mechanics T_0 data indicate a strong irradiation reaction even at higher fluence as shown in Figure 4.

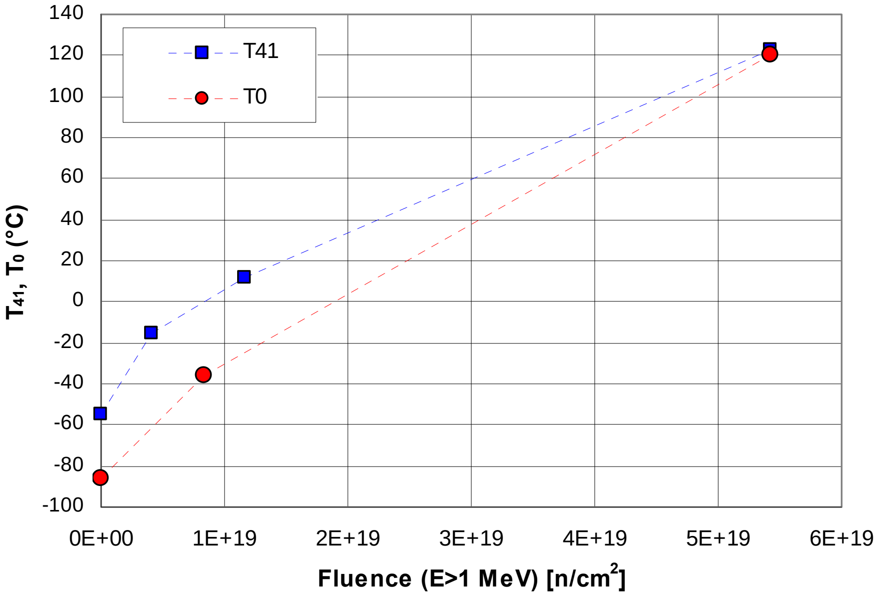

Figure 4: Measured T_{41} and T_0 data for P16 WM

For P16 WM weld it was revealed by APT analyses performed by Rolls-Royce that the number density, volume fraction and size of the irradiation-induced MnNiSiCu clusters increase steadily with fluence. As stated in [13] the number density increased from $2.2x10^{23}m^{-3}$ (at $0.41x10^{19}$ n/cm^2) to ~$6x10^{23}$ m^{-3} (at $1.16x10^{19}$ n/cm^2) to $16x10^{23}m^{-3}$ (at $5.22x10^{19}$ n/cm^2). The equivalent increase in volume fraction of the clusters was from 0.047 % to 0.21% to 2.1%. The correlation between square root of volume fraction and the shift in the Cv transition temperature is linear. Moreover, many of the MnNiSiCu features were associated with dislocations which were decorated by P as observed in all irradiated conditions [10]. The main effect of irradiation was a significant increase of the measured concentrations of Mn, Ni, Si and Cu (at the dislocations) as shown in Figure 5. This is consistent with the pinning of dislocations by MnNiSiCu clusters [10].

Again, the fluence depending change in material properties (T_{41}, T_0) could be confirmed by the measured increase of number density and volume fraction measured by APT.

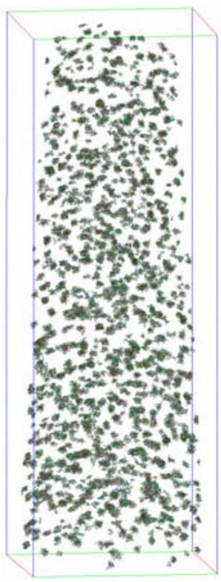

Figure 5: Mn/Ni/Si/Cu clusters in 1.69 % Ni weld P16 WM irradiated to $5.22x10^{19}$ cm^{-2} [13]

Low copper/nickel/phosphorus effect

For the material with the lowest Ni (0.84 %) and phosphorus (0.006 %) and low copper (0.05 %) contents P147 BM the measured T_{41} and T_0 data are shown in Figure 6. Obviously, this material was of the lowest irradiation-induced embrittlement compared to the other materials of the four investigated steels.

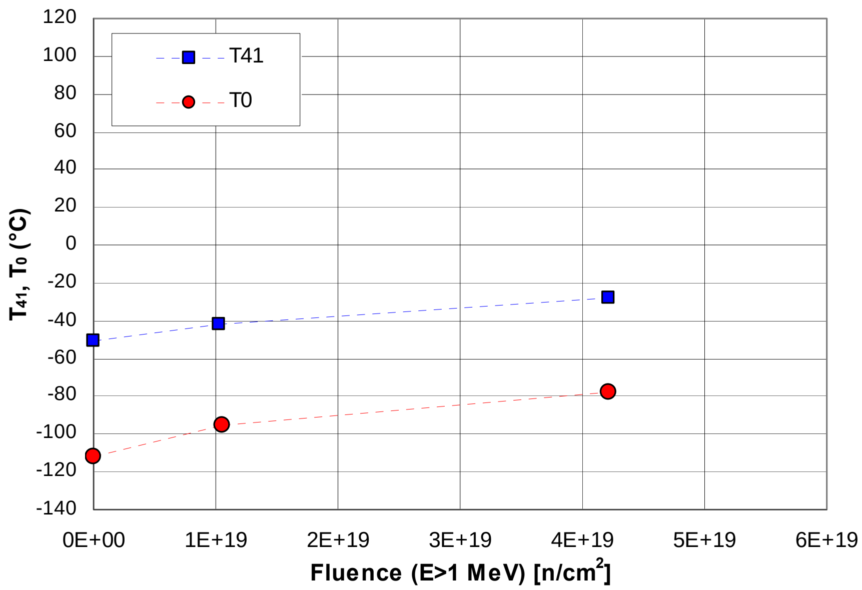

Figure 6: Measured T_{41} and T_0 data for P147 BM

The total volume fraction and number density of irradiation-induced features have been measured by SANS experiments performed by HZDR Dresden and are summarized in Table 7.

Even if a moderate increase of volume fraction and number density was observed, the absolute values of irradiated P147 BM are very low compared to the other three materials P141 WM, P370 WM and P16 WM. This indicates some matrix damage effects rather than any significant cluster formation for the highly irradiated P147 BM.

Table 7: Irradiation-induced clusters for P147 BM measured by SANS [10]

Material	Fluence, Φ (10^{19} cm^{-2}) (E>1MeV)	Volume fraction, c (vol%)	Number density, N (10^{16} cm^{-3})	Radius, R_{mean} (nm)	Radius, R_{peak} (nm)	A-ratio
P147 BM	1.05	0.008±0.005	4±3	0.74	0.50	1.5±0.2
	4.24	0.027±0.005	16±10	0.70	0.50	-

TEM imaging was carried out by CIEMAT under WBDF conditions (Figure 7, left). Dislocation loops, if present, would have the appearance of white small dots (depending on the size). As can be seen on the image no dislocation loops were observed throughout the sample [10]. Figure 7, right shows the development of the size distribution of solute clusters with neutron fluence, determined by SANS measurements.

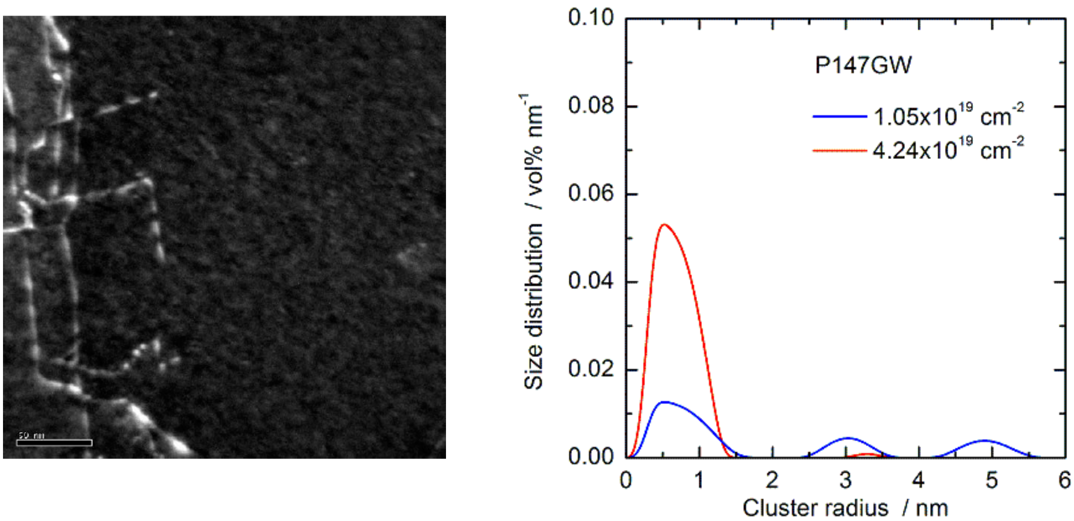

Figure 7: TEM image under WBDF conditions for material P147 BM (left) and reconstructed size distributions from SANS (right), for 4.22x10^{19} n/cm^2 [10]

Thus, the measured low increase of number density and volume fraction measured by SANS and the TEM data confirmed the low irradiation-induced embrittlement of the well optimized material P147 BM with comparatively low contents of Ni, P, and Cu.

Prediction by trend curves

The ΔT_{41} data measured for all four RPV steels have been compared with the following well known prediction formulas (trend curves) which are implemented in the ASTM E10.02 PLOTTER Tool:

- Reg. Guide 1.99 Rev. 2 Pos. 1 [14]
- ASTM E 900 - 02 [15]
- FIM Formula (Todeschini, EdF 900 MW) [16]
- Wide-Range Model WR-C(5) [17]
- Erickson CVE-Model [18]
- 10 CFR § 50.61a (EONY) [19], [20]
- JEAC4201-2007 [21]

For reasons of clarity the results of the trend curves described above are shown in Figure 8. A big scatter of the predicted results is obviously, however there is always at least one trend curve which is able to predict the measured data with sufficient reliability. Whereas the Reg. Guide 1.99 Rev. 2 Pos. 1, EONY and ASTM E900-02 models give good predictions for the low Cu/Ni materials P141 WM and P147 BM, the Erickson model seems to be more appropriate for the high copper weld P370 WM. For the high nickel weld P16 WM only the JEAC4201-2007 formula is able to predict the measured T_{41} shift satisfactorily. However, more data points are needed to confirm these conclusions.

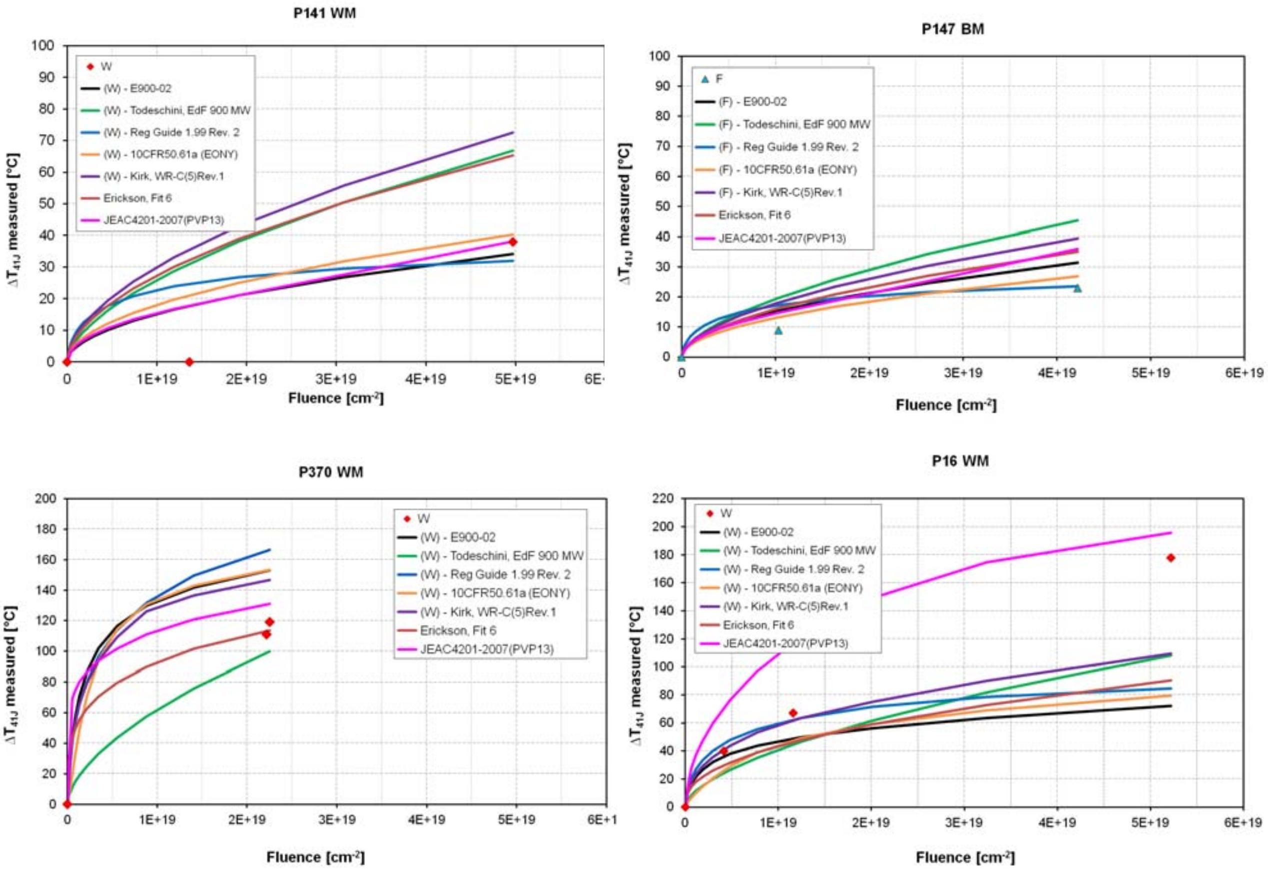

Figure 8: Measured ΔT_{41} data and predictions by trend curves

SUMMARY AND CONCLUSIONS

Four RPV steels which are particularly interesting under the point of view of long term irradiation behavior have been studied at different irradiation levels up to 8×10^{19} n/cm^2 (E > 1 MeV). The material properties in terms of T_{41} from Charpy testing and T_0 from fracture toughness testing were compared with data obtained by various microstructural analyses techniques such as APT, SANS and TEM. For all four materials the irradiation-induced change in material properties could be confirmed. Finally, the ΔT_{41} data measured for all four RPV steels have been compared with predictions from some well known trend curves.

In this way the understanding of the material behavior induced by neutron irradiation was improved and it was also shown that microstructural examinations may serve as a valuable assistance to the classical material testing in RPV irradiation surveillance programs.

With respect to best practices for RPV irradiation surveillance programs following consequences can be drawn from the study performed:

- Microstructural techniques such as APT, SANS, TEM are appropriate to investigate irradiation effects.

- If RPV materials with high contents of copper or nickel are monitored then an almost linear increase of irradiation induced toughness reduction may be expected with only a weak tendency of saturation if any.
- If any potential impact of neutron flux and lead factor respectively is assessed other sources of uncertainty such as different neutron spectra have to be taken into account.
- Late irradiation effects may occur occasionally, however reliable information is needed on the take out location of the specimens to exclude any significant material variability.
- Even if significant uncertainty may occur if prediction formulas are applied, it seems that there should be at least one trend curve which is able to predict the measured data with sufficient reliability. However, the challenge is to find the right one.
- For well optimized RPV materials with comparatively low contents of Ni, P, and Cu the small increase of irradiation induced toughness reduction can be predicted by most of the trend curves with acceptable margins.

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