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Reactor Pressure Vessel Task of Light Water Reactor Sustainability Program: M3LW-12OR0402012 - Letter Report on Metallurgical Examination of the High Fluence RPV Specimens From the Ringhals Nuclear Reactors

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Light Water Reactor Sustainability

Reactor Pressure Vessel Task of Light Water Reactor Sustainability Program: M3LW-12OR0402012 – Milestone Report on Metallurgical Examination of the High Fluence RPV Specimens From the Ringhals Nuclear Reactors in Sweden.

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1. INTRODUCTION

Regulations which govern the operation of commercial nuclear power plants require conservative margins of fracture toughness, both during normal operation and under accident scenarios. In the irradiated condition, the fracture toughness of the RPV may be severely degraded, with the degree of toughness loss dependent on the radiation sensitivity of the materials. As stated in previous progress reports, the available embrittlement predictive models, e.g. [1], and our present understanding of radiation damage are not fully quantitative, and do not treat all potentially significant variables and issues, particularly considering extension of operation to 80y.

The major issues regarding irradiation effects are discussed in [2, 3] and have also been discussed in previous progress and milestone reports. As noted previously, of the many significant issues discussed, the issue considered to have the most impact on the current regulatory process is that associated with effects of neutron irradiation on RPV steels at high fluence, for long irradiation times, and as affected by neutron flux. A significant issue associated with such predictive capability is that of RPV materials containing relatively high nickel content and the Ringhals reactors discussed in Section 2 of this report contain weld metals with high nickel contents. There are some U.S. RPVs with relatively high nickel content, e.g., the Palisades Nuclear Plant RPV contains a weld with about 1.3 wt% nickel. The primary objective of the LWRSP RPV task is to develop robust predictions of transition temperature shifts (TTS) at high fluence (ϕt) to at least 10^{20} n/cm^2 (>1 MeV) pertinent to plant operation of some pressurized water reactors (PWR) for 80 full power years. New and existing databases will be combined to support developing physically based models of TTS for high fluence-low flux ($\phi < 10^{11}$/n/cm^2-s) conditions, beyond the existing surveillance database, to neutron fluences of at least 1×10^{20} n/cm^2 (>1 MeV).

The strong synergistic interactions between copper, nickel and manganese are understood at low to intermediate fluence. However, the interactions at higher fluence, in both low and higher copper steels, needs to be established. Similarly the basic role of phosphorous is established. However, potential interactions with copper and phosphorous effects at high fluence have not been quantified. Similarly, Si-Ni and Si-Mn interactions also result in silicon enrichment in CRPs. Ref. [3] discusses this issue in some detail pointing out that, even in the absence of copper, thermodynamic-kinetic models predicted the formation of Mn-Ni phases, although at low nucleation rates compared to that for CRPs, the effect being that relatively high incubation fluences are required for their formation.[4-8] Figure 1.1 from Ref. [3] schematically illustrates in 1.1(a), however, that once nucleated such late blooming Mn-Ni-Si phases (LBP) rapidly grow to large volume fractions, potentially causing severe embrittlement. Moreover, the models also show that small concentrations of copper may act as a catalyst for such LBP nucleation. None of the current models for embrittlement reflect the potential LBP embrittlement contributions, probably in part because they may require critical combinations of higher nickel and fluence and lower temperature and flux that have yet to be extensively encountered in the surveillance database.[3] The existence of LBPs has been confirmed, as illustrated in Figure 1.1(b), showing an atom probe tomography (APT) map of manganese and nickel atom positions. An enlarged view of a Mn-Ni precipitate in a copper-free, 1.6Ni-1.6Mn wt.% model alloy is also shown.[9] Similar observations have been reported by other researchers around the world.

This report provides the status for the Milestone M3LW-12OR0402012 – “Provide letter report on metallurgical examination of the high fluence RPV specimens from the Ringhals nuclear reactors in Sweden.” This milestone is associated with procurement of material, preparation of specimens, microstructural examinations, and analysis of the results relative to current

understanding regarding irradiation-induced microstructural evolution in RPV materials.

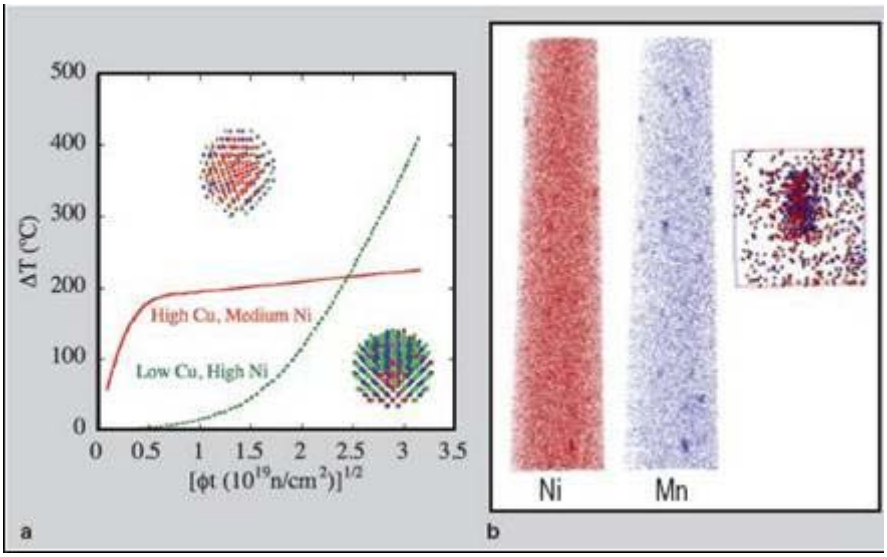

Figure 1.1. (a) Illustrative model predictions of the dose (in milli-displacements per atom) dependence of hardening in a high copper, medium nickel steel due to CRP hardening and in high-nickel, low-copper steel due to LBP hardening; (b) APT maps of nickel and manganese distributions and a blowup of a Mn-Ni LBP precipitate in a copper-free 1.6 wt.% Ni-1.6 wt.% Mn model alloy irradiated to 1.8x10^{19} n/cm^2 at high flux and 290°C. (From ref. 3)

2. BACKGROUND ON RINGHALS RPV MATERIALS

The following information is taken from Efsing, et. al. [10]. The Ringhals Units 3 and 4 reactors are pressurized water reactors (PWR) designed and supplied by Westinghouse Electric Company, with commercial operation in 1981 and 1983, respectively. The RPVs for both reactors were fabricated by the Uddcomb Company with ring forgings of SA 508 class 2 material made by Klöckner Werke. Surveillance blocks for both units were also supplied by Uddcomb using the same weld wire heat, welding procedures, and base metals used for the RPVs. The primary interest in these weld metals is because they are very high in nickel content, as seen in Table 2.1, with 1.58 and 1.66 wt% for Unit 3 and Unit 4, respectively. For reference, the highest nickel content in the U.S. Nuclear Regulatory Commission Regulatory Guide 1.99, Rev. 2 is 1.20 wt% [11]. As stated by Efsing, et. al. [10], the nickel content in Unit 4 is the highest reported nickel content for any Westinghouse PWR. This high nickel content is, as mentioned previously, the primary reason for interest in examination of the irradiated microstructure of the Ringhals welds.

As expected for such high nickel welds, both weld metals have exhibited very high irradiation-induced Charpy 41-J transition temperature shifts in surveillance testing. Figures 2.1 and 2.2, taken from [10], show results of surveillance tests from four capsules of Unit R3 and three capsules of Unit R4, respectively. Although the 41-J shifts and corresponding fluences are not indicated on the figures, ref. [10] provides the following detailed information:

1. For Unit R3, shift of 192°C (345°F) at 5.0×10^{19} n/cm^2 (>1 MeV)

2. For Unit R4, shift of 162°C (292°F) at 6.0×10^{19} n/cm^2 (>1 MeV)

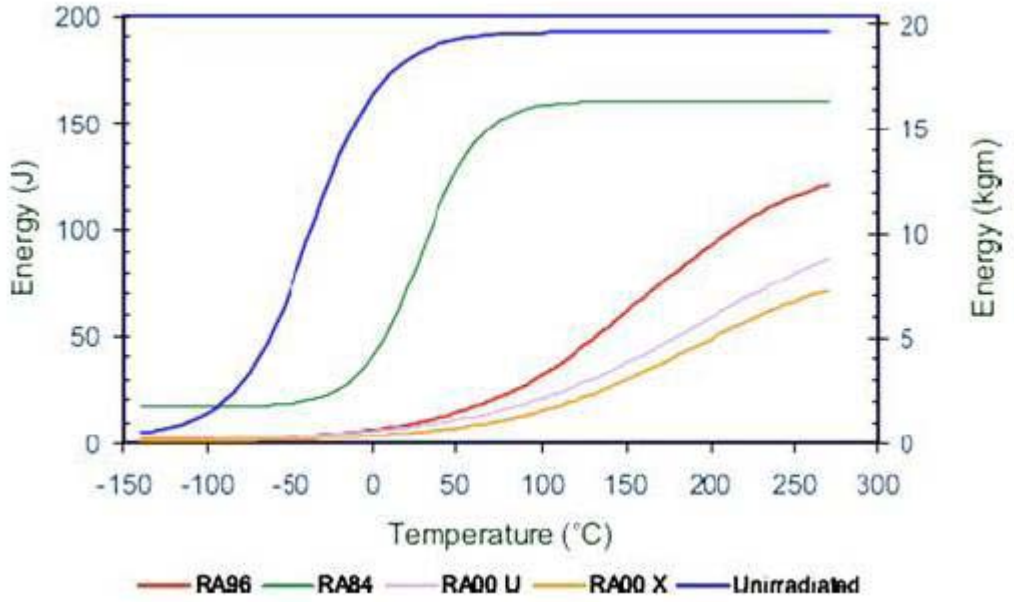

Figure 2.1 Summary of Charpy V impact testing of Ringhals 3 weld metal.

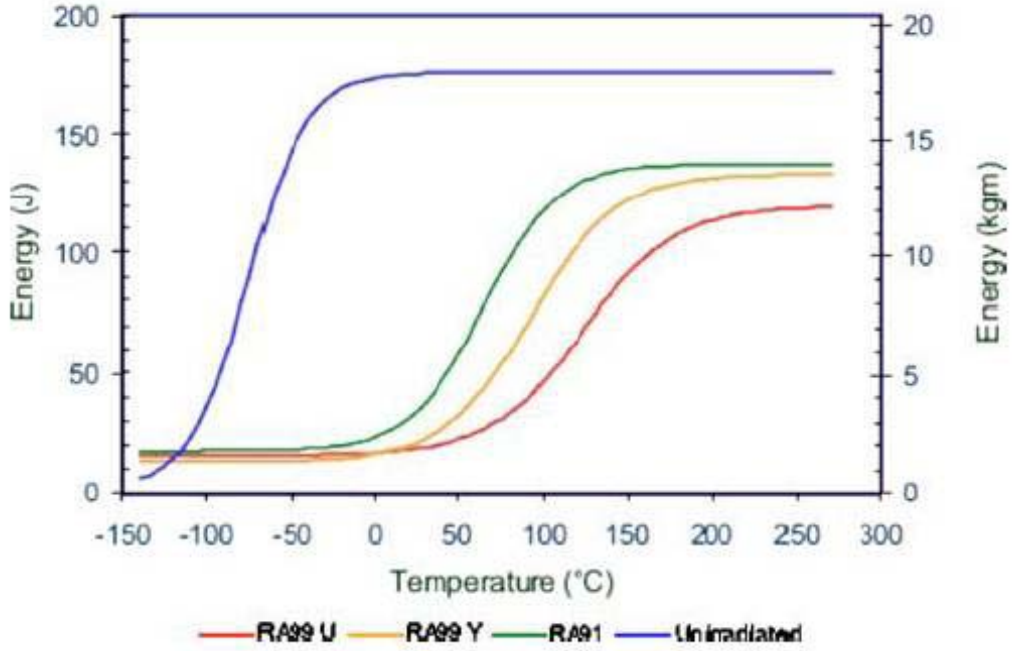

Figure 2.2. Summary of Charpy V impact testing of Ringhals 4 weld metal.

Those values of shifts do not correspond to the curves with the highest shifts in the figures, but the shifts provided in [10] are very high nonetheless. However, Figure 2.3, also from [10], shows the transition temperatures (at the Charpy energy of 41J) for both weld metals at the indicated

fluences. The two data points for the Unit R3 material in Fig. 2.3 indicate an average 41J temperature (T_{41J}) of about 170°C. Given a transition temperature of –68°C in the unirradiated condition for the Unit R3 weld metal, provided in [10], that would indicate a 41-J shift of 238°C (428°F) at a fluence of 6.8×10^{19} n/cm^2 (>1 MeV). Likewise, for the Unit R4 weld metal, the indicated shift at about the same fluence is 190°C (342°F).

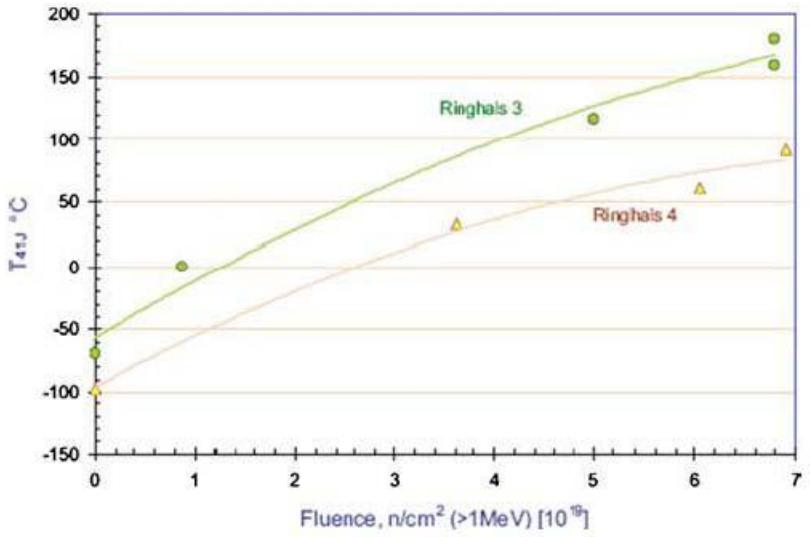

Figure 2.3. The ductile-to-brittle transition temperature (T_{41J}) as a function of fluence.

Thus, the radiation sensitivity of both those weld metals is extremely high in spite of their low copper contents of 0.08 and 0.05 wt% for R3 and R4, respectively. The RPV task requested surveillance material from Vattenfall AB, the operator of the Ringhals Nuclear Plants, for microstructural examination.

3. MATERIALS AVAILABLE FOR EVALUATION

Dr. Pal Efsing of Vattenfall AB in Sweden, has provided small samples of surveillance materials removed from tested Charpy impact specimens. All the surveillance specimens are from low-copper high-nickel weld metals in Ringhals Units R3 and R4. Table 3.1, from Efsing, provides the chemical composition for the materials, showing nickel contents of about 1.6 wt% and copper contents of 0.08 wt% or less. Table 3.2, also from Efsing, provides the irradiation temperature, neutron flux and fluence for each specimen. The samples provided by Vattenfall are slices from Charpy impact specimens and are about 10×10×0.4 mm thick. The objective of this task to use these specimens for preparation of samples for atom probe tomography (APT) and small-angle neutron scattering (SANS) to characterize the microstructure relative to irradiation-induced precipitates and other observable defects. As stated earlier, these materials are of high interest to the LWRS Program because they have low copper and high nickel contents, have been irradiated to relatively high neutron fluences, and exhibit very significant radiation sensitivity as indicated by the very high Charpy 41-J transition temperature shifts. This again relates to the issue of late-blooming nickel-manganese-silicon phases, especially since these materials have low copper contents.

Table 3.1. Chemical compositions of RPV surveillance materials from Ringhalls Units 3 and 4.

Matl	C	Si	Mn	P	S	Cr	Mo	Ni	Cu
Unit R3	0.052	0.21	1.46	0.009	0.006	0.07	0.54	1.58	0.08
Unit R4	0.068	0.14	1.35	0.0015	0.004	0.04	0.5	1.66	0.05
ASME 1971	<0.27	0.15-0.35	0.50-0.90	<0.025	<0.025	0.25-0.45	0.55-0.90	0.55-0.90	0.05

Table 3.2. Neutron exposure data for RPV surveillance materials from Ringhalls Units 3 and 4.

Specimen	Reactor	Capsule	Material	Temp, °C	Fluence	EFPY	Calculated Flux
E46,3A	R3	W	Weld	284	4.34E+19	10.4	1.32E+11
E54,3A	R3	W	Weld	284	4.34E+19	10.4	1.32E+11
E6,3A	R3	U	Weld	284	6.39E+19	13.8	1.47E+11
E63,3A	R3	X	Weld	284	6.39E+19	13.8	1.47E+11
E56,5	R3	X	Weld	284	6.39E+19	13.8	1.47E+11
E16,3B	R3	U	Weld	284	6.39E+19	13.8	1.47E+11
E56,3A	R3	X	Weld	284	6.39E+19	13.8	1.47E+11
N27,3B	R4	V	Weld	284	3.30E+19	6.3	1.66E+11
N27,3A	R4	V	Weld	284	3.30E+19	6.3	1.66E+11
N11,3A	R4	U	Weld	284	6.03E+19	12.8	1.49E+11
N18,3B	R4	U	Weld	284	6.03E+19	12.8	1.49E+11

To allow for adequate SANS analysis, we have requested unirradiated archive material for each weld metal as well. At the time of this report, atom probe blanks are being fabricated from the 0.4-mm thick samples. These blanks will then be electropolished and thinned into APT needles with APT measurements scheduled for the third week of April. SANS specimens of the irradiated materials have been prepared and are ready for measurements; the High-Flux Isotope Reactor (HFIR) had been shut down for some months which delayed the SANS measurements by some months, but the reactor is now operating. We are waiting for the neutron scattering experiment group at the HFIR to provide an experiment schedule for our specimens. Moreover, as mentioned, to complete the SANS analysis, we need to perform SANS measurements on the unirradiated material.

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