

Review

# Review on Steel Enhancement for Nuclear RPVs

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**Abstract:** The reactor pressure vessel (RPV) is one of the most important elements of a nuclear power plant (NPP). The RPV determines the plant operational lifetime since it is not replaceable economically. The purpose of the RPV steel study and enhancement to increase the NPP's (Nuclear Power Plants) operation lifetime from the original 30–40 years up to 60–80 years or even beyond. The RPV lifetime limited by ageing of the RPV steels. RPV ageing highly depends on the main environmental effects: fast neutron radiation, thermal effects causing thermal ageing and low-cycle fatigue. Firstly, the chemical composition via aged mechanical properties was studied. Efforts to increase the toughness against the radiation embrittlement was enhanced by the appearance of the modern microstructural testing devices such as APFIM (atom probe field ion microscopy), SANS (small-angle neutron scattering) positron annihilation spectroscopy (PAS), transmission electron microscopy (TEM) and Mössbauer spectroscopy (MS). The information on the effect of alloying and polluting elements for the microstructure allowed us to produce increased ageing toughness of the RPVs, and to enhance the safety and lifetime calculations of them, supporting long-term safe operation (LTO).

**Keywords:** reactor pressure vessel; radiation embrittlement; role of alloying and polluting element; long-term ageing



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## 1. Introduction

The role of the RPV is to keep the fuel elements, the core control devices and the coolant, as well as it being the most important part of the safety system between people and the radioactive core. The RPV determines the plant operational lifetime since it is not replaceable economically. During the last 60 years since NPPs have been operating, the safety requirements have been more severe despite no serious failure of RPV, and no RPV integrity problem has played a role in the NPP disasters which happened during the 60 years while the collected operational years exceeded 2000. The design requirement of the RPVs is that it should survive any operational malfunctions or accidental events of which can be assumed to occur during hundred thousand operational years.

The majority of the operating nuclear power plants are pressurized water reactors (PWR). This type of reactor is safe and reliable. The literature distinguishes the western PWR reactors and the Russian (soviet) WWER-reactors. This is rather political than technical, the WWER reactors are also pressurized water reactors, as well as the basic design of the two types of NPP, with the ageing mechanisms and design requirements being similar. Consequently, they will be discussed together in this paper.

The operating RPVs can be divided into three generations. The first generation was made from ordinary low-alloyed boiler steels. The purpose of the alloying was to increase the strengths and reduce the wall thickness. The design was based on the boiler design rules. The vessels were made from hot-rolled and bent thick plates welded together, including horizontal and vertical manual arc welds. The quality control of the vessels was also the same as for the welded boilers: a visual check, the liquid penetration crack test, X-ray, and the hydro pressure test. As thicker and thicker walled pressure vessels were produced, failures occurred during cold hydrotesting. The reason was the brittle weldment

of the thick-wall vessels. In thick structures, three axis tensile stresses occur around any defect. Three-dimension tensile stresses constrain the plastic deformations required to reduce the stress intensity at the defect tip. The development of the RPV steels and welding quality, using forged rings instead of plates, and the use of the elevated temperature hydro test and fracture mechanical failure analyses during the design process became common practice at the production of the second generation RPVs. The third generation of the RPVs are built from refined steels with limited polluting elements, and designed for 60 years of operating lifetime.

## 2. Materials and Methods

This review is based on a literature study as well as on experience collected during several European Framework research projects such as PISA [1], LONGLIFE [2], STRUMAT-LTO [3]. Most efforts until now were performed on the development of the pressurized and boiling water reactor vessel materials, but the collected experiences help the development of materials for other types of vessels, including different fourth generation fission and fusion reactors. These ones have to resist against high temperatures and corrosion, too. Specific requirements of the materials for these future designs are too wide to be summarized in one paper; consequently, this paper is limited to describe the enhancement of the materials of pressurized water reactor vessels.

Only a few types of steels are used for the RPV walls (see Table 1). Nuclear codes require very expensive and time-consuming testing programs to accept new structural material for RPV production. In many cases, the same type of steel has different names according to the manufacturer, but in the table only the standard names are used.

**Table 1.** The chemical composition of low alloyed steels used for pressurized water reactor pressure vessels.

		C%	Si%	Mn%	P%	S%	Ni%	Cr%	Mo%	V%	Cu%
SA508	min	0.15	0.15	0.66	0.006		0.37	0.31	0.37	-	0.03
	max	0.21	0.3	1.59	0.012		0.85	0.45	0.68	0.01	0.10
16MND5	min	0.15	0.16	1.21	0.003	0.001	0.68	0.15	0.47	<0.001	0.04
	max	0.18	0.34	1.50	0.012	0.016	0.80	0.28	0.56	0.025	0.09
22NiMoCr37	min	0.17	0.15	0.50			0.60	0.30	0.50		
	max	0.25	0.35	1.00	0.02	0.02	1.20	0.50	0.80	0.03	0.18
ASTM A-20/20M	min	0.19	0.22	1.32	0.006		0.56	0.03	0.51	0.01	0.02
	max	0.26	0.27	1.56	0.011		0.61	0.16	0.67	0.05	0.06
20MnMoNi-55	min	0.17		1.00			0.40		0.45		
	max	0.23	0.35	1.50	0.020	0.020	0.8	0.3	0.60		0.18
SA-302 B	min		0.13	1.07	-	-	-		0.41		
	max	0.25	0.45	1.62	0.035	0.035	-		0.64		
SA533B	min	-	0.15	0.8	-	-	0.37	-	0.35	-	-
	max	0.25	0.60	1.8	0.035	0.035	0.85	-	0.65	-	-
16Mo 5	min	0.12	0.15	0.50	-	-	-	-	0.45	-	-
	max	0.20	0.50	0.80	0.04	0.04	-	-	0.65	-	-
15Kh2MFA	min	0.13	0.17	0.30				2.50	0.60	0.25	
	max	0.18	0.37	0.60	0.025	0.025	0.40	3.00	0.80	0.35	
15Kh2NMFA	min	0.13	0.17	0.30			1.00	1.80	0.50		
	max	0.18	0.37	0.60	0.020	0.020	1.50	2.30	0.70	0.10	

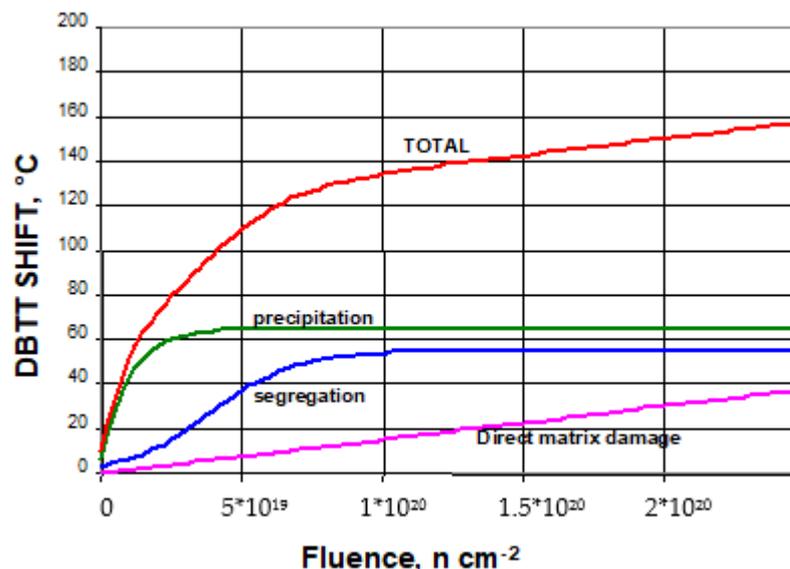
Many standards do not limit the pollution element, such as P, Cu, etc. These case the designer or manufacturer prescribe the allowable maximum quantity.

The residual elements, especially copper and phosphorus, and nickel as an alloying element, play an important role in irradiation hardening and embrittlement. However, until recently, it was not possible to clearly identify the irradiation-induced changes in the microstructure. Progress was made in microstructure examination using techniques such as APFIM (atom probe field microscope), SANS (small-angle neutron scattering), PAS (positron annihilation spectroscopy), and TEM (transmission electron microscopy). They allowed us to identify the irradiation induced defects, segregations and precipitations, and consequently, the effects of the alloying elements became more clear, and synergetic effects are considered.

### 3. Irradiation and Thermal Ageing of RPV Steels

The operation conditions of the RPV determine the required properties of the RPV steels. The main required properties: toughness against fast-neutron irradiation and thermal ageing, low activation, good weldability and machineability, and high strength. To satisfy all of these requirements, carefully designed alloying is necessary.

The most severe requirement for the RPV steels is the irradiation toughness. Neutron irradiation causes three types of damage in the structural materials: precipitates and segregations occur, and the lattice can be damaged. The sum of the effect of these three mechanisms gave the embrittlement, as can be seen in Figure 1 [4]. The total value describes the transition temperature increase in the function of the fluence (neutron irradiation dose). This curve determines the lifetime of the pressure vessel. If one element initiating segregation clusters is depleted in the matrix, other ones may initiate new types of segregations. Copper precipitations are the first, but later Ni–Mn precipitates became the major players. This is called late blooming, since the second type of precipitates may accelerate the embrittlement and deform the total curve in Figure 1. [5]. If the DBTT shift reaches a critical value (calculated for each unit) the vessel safety level becomes too low to operate it.

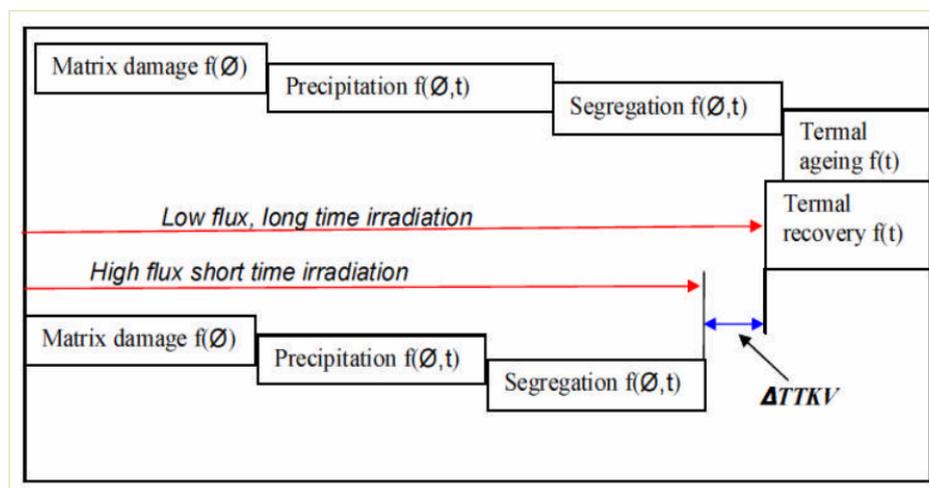


**Figure 1.** The processes of the radiation embrittlement. Direct matrix damage depends on the displaced atoms, and until large fluences, it is rational with the fast neutron fluence [4]. The precipitations and segregations occur at the beginning of the irradiations and saturating at medium fluence values because the low displacement tough elements move into the segregations and precipitations and become depleted in the matrix.

The second important degradation mechanism is thermal ageing. The PWR's operating temperature is in the range of 270–320 °C. For steels, this temperature is too low to cause serious degradation of the material properties; however, thermal ageing is a diffusion process and neutron irradiation may accelerate it. Analyses of surveillance (specimen sets aged in the operating reactor vessels) databases show that only nickel-alloyed steel ( $\text{Ni}\% \geq 1.5\%$ ) shows considerable thermal ageing, and in other steels no degradation is detected or only so small an amount that it is shadowed by the scatter of the mechanical testing. Sometimes, surveillance results even show a limited negative thermal ageing effect (annealing) [6].

The rate of degradation depends on the environmental conditions as fluence, temperature, neutron flux and neutron spectrum.

Figure 2 shows how the neutron flux affects the ageing. In a large flux, the time to reach a certain fluence is short, and consequently, less time is available for diffusion. As a result, the thermal part of the embrittlement is shorter and results in less ageing. This is the reason that the measured degradation in a high-flux material testing reactor is smaller than in long-time surveillance tests.



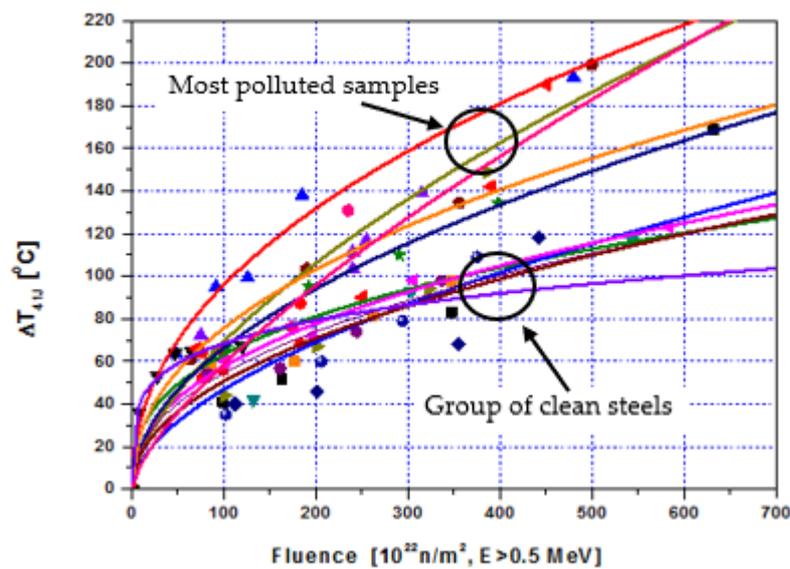
**Figure 2.** Summary of the RPV ageing mechanisms and comparison of high- and low-flux irradiation at low temperature ( $\leq 350$  °C). At high-flux irradiation (in accelerated research facilities) the time is short for finishing the diffusion processes, and the role of thermal ageing and thermal recovery (annealing) is negligible. At higher temperature, the diffusion accelerated, and thermal annealing may reduce the radiation embrittlement [7].

The third degradation mechanism is the low-cycle fatigue. At the start and shut down or at a load change, thermal stresses occur in the thick reactor wall. The RPV inside is covered by a welded austenitic layer (cladding) to avoid corrosion. However, the thermal expansion coefficient of this layer is nearly double that of the low alloyed RPV steel, indicating thermal stresses, and can initiate low-cycle fatigue cracks at the border of the two types of material. Low cycle thermal fatigue in the bulk RPV materials is not typical. To avoid the low-cycle fatigue damage of the intermediate zone, the designers limit the number of operational cycles (shut down and restart, quick- and high-load change) during the operational life. Periodical in-service inspections (ultrasonic and magnetic testing) of the transition zone between the austenitic cladding and the vessel low-alloyed steel is the tool to avoid low-cycle fatigue crack initiation.

The material toughness against these ageing factors determines the lifetime of the RPV, and the main direction of the development of the RPV steels is to slow down the above-mentioned ageing processes to ensure long-time operation. Originally, the RPV licences were given for 30 or 40 years; nowadays, many efforts going on the extend it beyond 60 years.

#### 4. The Chemical Composition Effect on Radiation Embrittlement

The chemical composition (alloying and polluting elements) is the main factor which determines the radiation embrittlement. Figure 3 shows an example of how much a difference in radiation embrittlement can occur within a steel type produced according to the same standard [7]. Different batches of the 15Kh2MFA steel show about a 50% difference in the DBTT (ductile brittle temperature transition measured with Charpy impact testing using the 41 Joule criteria). The effect of most typical alloying and polluting elements is discussed below.



**Figure 3.** 15Kh2MFA forging transition temperature change in the function of the neutron fluence. It shows the effect of the chemical composition: the ductile brittle transition temperature reduced (measured by Charpy impact test using 41J criteria) decreased by about 50% using clean version of the steel [7].

##### 4.1. The Effect of Copper

Copper is not an alloying element in RPV steels, but it is present as a residual when Cu-containing scrap has been used in the production of the original ingot, or when Cu has been used in wire welding coating. Despite the low levels of this element (very rarely as much as 0.5% even in older welds), it seriously affects the irradiation sensitivity of the steel [8]. It was concluded that the Cu content should be kept as low as possible, typically below 0.1% to decrease the irradiation embrittlement [9].

Extensive research experiments were carried out to quantitatively define the effect of Cu, and other residual elements in the IAEA CRP-3 program [10] and in the HSST program [11] in the U.S. After establishing the detrimental effect of Cu on the mechanical properties, an in-depth microstructural examination started to identify the underlying defects responsible for the change in the mechanical properties. The role the Cu is to develop point-defect clusters. Vacancies interact with small Cu clusters to form defect aggregates which are more stable and more numerous than those ordinarily formed in the absence of Cu. The clusters of Cu atoms serve as nucleation sites for defect aggregates [8]. It was discovered that even at <0.1 wt%, Cu was present above its solubility limit, and at RPV operating temperatures Cu-based clusters occurred [12]. Initially the precipitates were expected to be pure Cu, but microstructural analyses showed a more complex picture [13]. The use of APFIM, SANS, PAS and high-resolution TEM show that Cu- and P-rich regions (clusters) occur as an effect of neutron irradiation [14]. Other alloying and polluting atoms may join to these clusters [15].

In addition to its effect on increasing the irradiation sensitivity of steel, there are indications that the high Cu content in RPV steels retards the recovery of the mechanical properties during post-irradiation annealing process [16].

The study of the irradiation effect on 15Kh2MFA steel concluded that by hardening the single copper role, it is relatively low compared with the effect of phosphorus and another mechanism. In contrast the synergetic effect of copper and phosphorus is a main contributor [15].

When precipitation is thermally induced, the nucleation and growth stages are followed by coarsening, and the larger precipitates grow at the expense of the smaller ones. Coarsening, or ripening, usually begins before precipitation has reached completion, and results in softening, or over-ageing. Over-ageing is not typical in PWRs, the reason of it is the presence of other alloying elements. The softening caused by the depletion of the alloying elements in the matrix are less than the hardening caused by the precipitates and the increased number of dislocations. Over-ageing may start at a higher irradiation than the usual lifetime fluence of the PWRs.

#### 4.2. The Effect of Phosphorous

The effect of P content on irradiation embrittlement was defined by U.S. NRC Regulatory Guide 1.99 Rev.1 [16] by including a “P term” in the formula used to determine the change of the ductile-to-brittle transition temperature (DBTT):

$$\Delta\text{DBTT} = (5/9) \times [40 + 1000 \times (\text{Cu}\% - 0.08) + 5000 \times (\text{P}\% - 0.008)] \times \sqrt{(\Theta / 10^{19})}$$

where  $\Theta$  is the fluence at E (neutron energy)  $\geq 1$  MeV.

The effect of P was greatest when Cu content is low (<0.1%). The sensitivity of the changes of the mechanical properties caused by the P content reduced as the Cu content raised. At  $\text{Cu} \leq 0.1$  wt%, research confirmed the significant effect of P-content over 0.003 wt% [16]. The average increase in the transition temperature was 20–30 °C for each 0.01 wt% of P for a Cu content of about 0.05 wt% and neutron fluence  $0.5\text{--}7.10^{19}$  n/cm<sup>2</sup>  $E \geq 1$  MeV.

APFIM examinations showed that phosphorous was found to form several types of clusters depending on the chemical composition of the steel. P clusters were often enriched with nickel and occasionally with carbon [17]. Generally, in commercial RPV steels, these features are present in a much lower density than the Cu clusters. In cases where the Cu level was very low or where the P content was increased, the density of the P clusters became significant. The density of the P clusters increased with increasing the P content while their average radius remained nearly constant at 0.5–1 nm. In contrast, when the Cu content was raised to 0.3 wt%, the clusters were Cu-rich clusters, and the P addition led to a refinement of the defect microstructure.

The observation of P clusters in irradiated steels led to the conclusion that the effect of P was caused by precipitation hardening and the depletion of P in the solid solution.

The effect of P on irradiation embrittlement also connected to the segregation of P at interfaces as grain boundaries and increased by the radiation enhancement of diffusion. P segregation weakens interfaces, causing intergranular brittle fracture (temper embrittlement), and microvoid nucleation around small precipitates.

P effects on embrittlement during irradiation were studied extensively during the EU-funded 5th Framework Project “PISA” [1]. P segregation at the grain boundaries was found to increase linearly with the dose, with little or no effect of the dose rate or irradiation temperature. The study of surveillance databases showed no evidence for non-hardening embrittlement. The reason is that most of the P remained in the Cu-based clusters, and the end of life fluence of the operating RPVs is low to allow a dangerous P layer at the grain boundaries.

A very interesting result shows that the distribution of P is not homogenous in the weld, in the middle section the P content is very high, and near the surface, the P content is low.

If P segregation is dominant, then non-hardening embrittlement could occur. The PISA program suggested that the rate of P segregation was low in the MnMoNi, CrMo and CrMoV steels. However, it is expected that hardening encourages intergranular failure at a given grain boundary P level [1].

#### 4.3. The Effect of Nickel

The contribution of Ni-content to irradiation hardening and embrittlement was realised well after the effect of Cu. It was shown that this effect is important for steels having Ni contents greater than 0.4 wt% [18]. Materials with high Cu and Ni contents will have high-irradiation sensitivity while those with low Cu and Ni content will be much less sensitive to irradiation [18].

Most of the published data show that the Ni effect depends on Cu content, and Ni content up to 1.2 wt% does not appear to play any role when the Cu content is low (<0.1 wt%). This suggests the existence of a synergetic mechanism resulting from interactions between Cu and Ni [19]. For Cu contents less than 0.1 wt%, the Ni effect was experienced only at high fluences. The synergetic effect was assumed to be a result of the participation of Ni in Cu-rich clusters formed during irradiation.

Microstructure examination showed that the enhancement of hardening with increasing Ni content in RPV steel was accompanied with the reduction in the mean size of Cu-rich clusters and an increase in cluster density. Cu-rich atmospheres and clusters, induced by irradiation, contain significant Ni enrichment. The calculation of Ni and Mn concentrations in the Cu-rich phases suggested that the combined concentration of Mn and Ni in the Cu-rich phases in irradiated RPV steels was 30% or more. Cu–Ni-rich clusters are formed in FeCuNi ternary alloys. The Ni/Cu ratio was 1:3 compared to 1:50 for the particles found before irradiation. Nickel was also found to be associated with P-rich clusters.

Nickel affects both the size (decreases) and distribution (more homogeneous) of the Cu-rich clusters (or precipitates). Because these clusters are supposed to act as obstacles to the dislocation motion resulting in hardening and embrittlement, Ni can contribute to irradiation hardening and embrittlement through influencing and participating in these clusters. Studies of RPV steels in surveillance programmes confirmed the presence of clusters rich in Ni, Mn and Si formed after irradiation [19]. The formation of solute clusters during the irradiation of RPV steels resulted in the depletion of Cu and P in the matrix, but investigation showed that Ni retards this depletion.

Increasing the Ni content of a Cu-containing steel produces a higher level of precipitation hardening at a given dose, but Ni appears to suppress over-ageing.

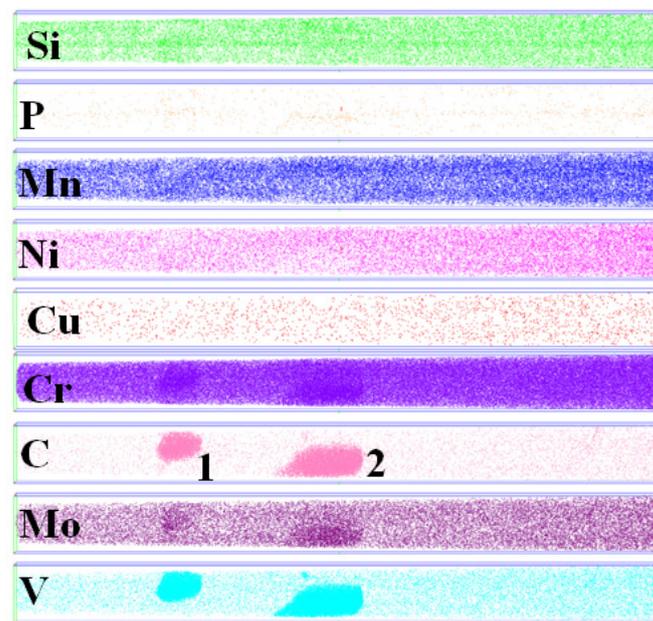
Nickel in the low alloyed steels causes thermal ageing, and the low temperature irradiation accelerates the thermal ageing [20].

In summary, Nickel increases the sensitivity for thermal ageing and radiation embrittlement shift but reduces the original transition temperature shift. The combination of these two effects limits the Ni content (0.4–0.5%) to provide an optimum for LTO.

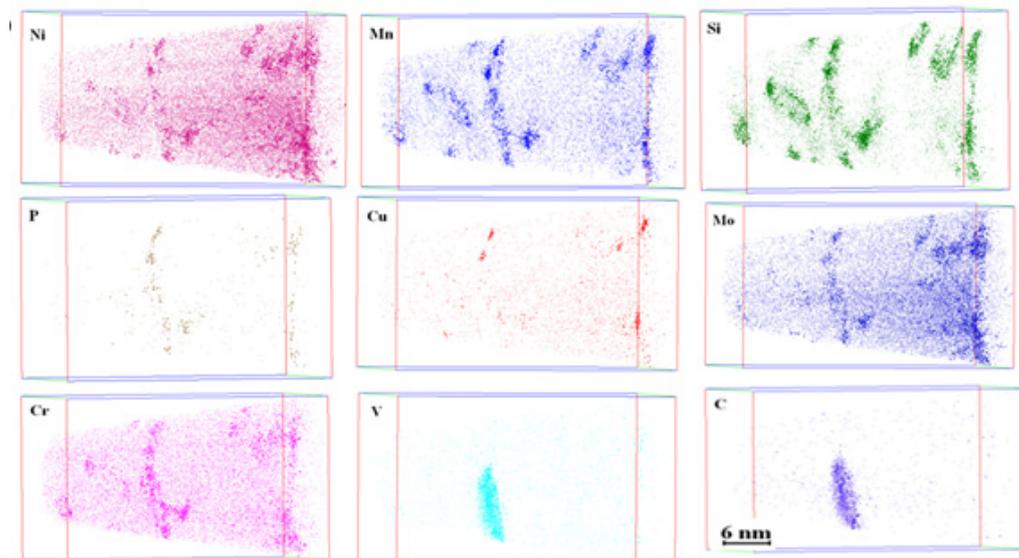
#### 4.4. The Effect of Mn and Ni

In RPV steels, Mn is incorporated into copper enriched clusters and enhances the incorporation of Ni into the precipitates [21].

Low-Cu or Cu-free solute clusters incorporating Mn, Ni and Si have also been observed in RPV steels. PAS, APT, SANS and TEM indicate that Mn plays a dominant role in the first stage of the formation process, before the contribution of Ni becomes more and more important. Figures 4 and 5 show the microstructure of a low copper Cr–Mo–V RPV steel in a received and highly irradiated condition. It can be seen that the initiation distribution (Figure 4) is nearly homogeneous, apart from the area of large carbide precipitations. After irradiation, few Cu segregations can be seen (Figure 5); however, the majority of the irradiation-induced clusters are not Cu but Mn initiated, and several other pollution and alloying atoms joined them.



**Figure 4.** APFIM results of as received Cr–Mo–V steel. The phosphorus, copper and other alloying and polluting elements are distributed in the matrix, only around vanadium carbides the chromium and molybdenum enriched slightly.



**Figure 5.** APFIM results of highly irradiated ( $6.19 \times 10^{20} \text{ n/cm}^2$ ,  $E > 1 \text{ MeV}$ ) Cr–Mo–V steel. Stronger colors mean increased density. Cu precipitations occur first and the other elements join to it forming clusters. Vanadium carbides occurred still in the foundry, but during irradiation other alloying and polluting elements join to them. It is also can be seen that that many clusters are not Cu but Mn based.

In low Cu RPV steels, the Mn–Ni clusters occur when the Cu content runs out in the matrix, and accelerates the embrittlement after a threshold fluence. This mechanism is called late blooming. The existence of late blooming in RPV steels is widely discussed since the effect may be covered up by the testing scatter. LTO is increasing the importance of the further study of the combined effect of Mn–Ni clusters and late blooming. The European framework project “STRUMAT-LTO” [3] is going on to study this effect.

#### 4.5. The Effect of Vanadium

The RPV wall material of the WWER reactors (15Kh2MFA) is alloyed with Cr, Mo and V. The vanadium alloying is expected to increase the radiation toughness, but there is no proof of it in the literature. The WWER-440 reactors suffer the largest EOL fluence among the PWRs operated in Europe, and the irradiation induced transition temperature shift is small compared with the other steels having similar Cu and P contents [22]. Phosphorus segregation to vanadium carbonitride interfaces was observed [23].

#### 4.6. The Effect of Other Polluting Elements

Very little information exists on the effect of other polluting elements on the radiation embrittlement. Most probable is the reason that follows: the RPV steels are very clean materials, having limited polluting elements. Most polluting elements are in the range of 0.01%. Thus it is difficult to separate the effect from the main components. The literature deals with the effect of some polluting elements. It stated that boron, tin, and stibium has effect on the radiation embrittlement; in contrary, the limited amount of sulphur, bismuth and lead do not change the radiation embrittlement shift. However, they may decrease the initial toughness properties [3].

### 5. Future Reactor Steels

This paper discussed the second- and third-generation RPV steels and their development, since today these reactors give the majority of nuclear electricity. Molten salt and molten metal, high temperature gas and supercritical water-cooled reactors are planned to be developed for the future. Presently, there are two main directions of the research involving the development of RPV steels:

- The elaboration of neutron radiation tough ferrite-martensitic steels with reduced activation (RAFM). These steels are alloyed with 9–14% Cr and contain some percent of other low activation elements;
- Another way to use oxide dispersed steels for high temperature vessels, but these steels are still in the research laboratories, and the industrial application of them is not allowed yet.

The future purposes of the engineers are to increase the fuel economy, to reduce the heat loss, and to burn the actinides (highly radioactive elements in the burn fuel). All of them need new structural materials resistant against corrosion, a high temperature, and at least a one-magnitude higher neutron dose than the life exposition of the present operating RPV.

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