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Intergranular and intragranular phosphorus segregation in Russian pressure vessel steels due to neutron irradiation

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Abstract

Russian reactor pressure vessel steels have been studied in three conditions: initial, irradiated and annealed. It has been established that irradiation induces both intergranular as well as intragranular phosphorus segregation. Fractographic studies demonstrated that brittle intergranular and ductile intergranular fracture surfaces of Charpy specimens appear as a result of intergranular and intragranular segregation, respectively. Transmission electron microscope (TEM) studies have revealed radiation-induced precipitates on interface boundaries to which intragranular phosphorus segregation occurs. Auger electron spectroscopy (AES) has been applied to detect phosphorus enrichment of fracture surfaces in the regions of brittle and ductile intergranular fractures. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

It is well known that radiation embrittlement (RE) of reactor pressure vessel steels (RPVS) induces the shift observed in transition curves obtained using Charpy tests defined as an increase in DBTT [1,2].

In addition to hardening, a significant contribution to RE can be brought about by intragranular segregation of impurities (primarily, phosphorus), occurring at interface boundaries of precipitates (including those arising under irradiation for, e.g., copper-enriched or copper-vacancy clusters) [3]. Certain contributions to RE in RPVS can be brought about by intergranular segregation – for, e.g., phosphorus [3].

Many studies devoted to grain-boundary phosphorus segregation in steels (the so-called phenomenon of 'reversible temper brittleness') are available [4,5]. In contrast, the number of studies, where intragranular phosphorus segregation was considered, is much less. However, in these few studies, the existence of phosphorus segregation at interface boundaries of the precipitate/matrix type together with an increase in DBTT induced by intragranular phosphorus segregation in steels was demonstrated using direct experimental methods. In particular, in [6] such segregation was observed in RPVS A533, A508 and in some model steels. It should be noted that intragranular segregation in these alloys was due to thermal aging at various temperatures. In [7] three different mechanisms of DBTT shift, i.e., hardening, formation of grain-boundary and intragranular phosphorus segregation, contributing to embrittlement occurring in steels subjected to thermal aging, were experimentally separated. In [6,7] it was shown that in the RPVS and model steels studied, intragranular phosphorus segregation proceeds mainly on M_6C type carbides, Laves phases and non-metallic inclusions. It was shown in the same studies that if any of the above types of precipitates are located along grain boundaries, then phosphorus segregation at their interfaces can lead to the appearance of ductile intergranular fractures on the surfaces of Charpy specimens tested at temperatures in the upper shelf (US).

The phenomena related to intragranular segregation were found in investigations of RE in some model binary alloys namely Fe–P and Fe–Sn in [8]. In this paper, the influence of concentration of the second element in

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the series of binary alloys, Fe–Ta, Fe–W, Fe–Nb, Fe–Ti, Fe–Cu, Fe–P, Fe–Sn, on the value of their RE resulting from irradiation to neutron fluences (3–5) × 10^{19} cm^{-2} ($E > 0.5$ MeV) at 50°C was investigated. Moreover, it was demonstrated that for all binary alloys (including the alloys Fe–Cu), but excluding Fe–P and Fe–Sn, the concentration dependencies of the yield stress after irradiation have the same shape as the concentration dependencies of the DBTT. It should be noted that for binary alloys Fe–P and Fe–Sn this relation does not exist. It should be emphasized as well that in Charpy specimens for alloys Fe–P and Fe–Sn the regions with intergranular fractures were not observed. As the authors [8] believe, this fact proves the dominant role of intragranular segregation in RE proceeding in these binary alloys.

In [6,7] the methods of FEGSTEM and APFIM were used to study intragranular phosphorus segregation at interface boundaries for Russian and American grade steels irradiated in conditions characteristic of an operating PWR.

Thus, the accumulated experimental data at the present time on investigation of RPVS show that in addition to hardening, irradiation can result in grain boundary and intragranular segregation of impurities (primarily, phosphorus).

It would be reasonable to suppose that the fractography of Charpy specimens tested at different and comparable temperatures before and after irradiation can provide direct information on the mechanisms responsible for RE in RPVS. As it is well known, in the majority of cases, for steels with bcc crystal lattice (in particular, for RPVS) schedules of final heat treatment are applied in order to prevent the occurrence of temper brittleness. For this reason, the fracture surfaces of unirradiated RPVS specimens represent various combinations of ductile cleavage and quasi-cleavage fracture [9,10]. In the simplest case, if RE in RPVS is caused only by hardening, then irradiation does not induce the appearance of new types of fracture modes. In the real case, when RE in RPVS is caused by the simultaneous action of several mechanisms, including the formation of grain boundary and intragranular segregation of impurities under irradiation, the fracture surfaces may also contain regions with brittle and ductile intergranular fractures [3,10]. It is unlikely that a comprehensive evaluation of the mechanisms responsible for RE in RPVS can be achieved without structural investigation of the same materials after recovery annealing and re-irradiation.

Therefore, in the present study a series of fractographic, structural and Auger electron spectroscopy (AES) investigations of RPVS in the initial, irradiated, annealed and re-irradiated states have been performed. The objective of the study was to provide a comparative analysis of all the investigations to achieve detailed information on the conditions of grain boundary and intragranular phosphorus segregation and their role in RE occurring in RPVS.

2. Materials and experimental

The following steels have been investigated:

- 15Kh2MFA – base metal (WWER-440). After forging, heat treatment: quenching 1000°C/10 h; cooling in oil; tempering 700°C/16 h, cooling in air.
- 15Kh2NMFAA – base metal (WWER-1000). After forging, heat treatment: austenisation 920°C/h, cooling in water; annealing at 650°C, cooling in air; annealing at 620°C/25 h, annealing at 650°C/20 h, final cooling in a furnace to room temperature.
- 25Kh3NM – base metal (prototype reactor). The material was subjected to heating to 870–890°C, air cooling to 700–800°C, cooling in oil; tempering at 620–670°C, subsequent cooling in a furnace.
- SV-10KhMFT – weld metal (WWER-440). Post-weld heat treatment: holding at 665°C (15 h), furnace cooling to 300°C, then air cooling.

The chemical composition of these RPVS is presented in Table 1.

The microstructure of the above base metal alloys consists of tempered bainite. Excess ferrite precipitates are also present in Russian weld metals in addition to bainite.

Embrittlement in irradiated specimens has been evaluated using DBTT shift and also reduction in the level of US as measured using standard Charpy specimens with a V-shape notch.

Fractographic investigations have been carried out on the halves of Charpy specimens chosen and stored under vacuum just after testing to preserve the fracture surfaces.

The fracture surfaces were examined using an X-ray microbeam analyser type SXR-50 – radioactive version (Cameca, France) installed in a hot cell. The images of the fractures were obtained with secondary electrons at an accelerating voltage of 20 kV and a probe current of 0.8 nA with a magnification in the range 50–3500×. The proportions of different types of fracture (ductile, brittle intercrystalline, ductile intercrystalline, cleavage and quasi-cleavage) in the total fracture surface after testing at different temperatures were evaluated using the Glagolev method [11]. The absolute error of measurements at 95% confidence level did not exceed 5%. Testing temperatures for each material corresponded to US, DBTT and lower shelf in the temperature dependence of ductile toughness.

An electron microscope type TEMSCAN-200CX was used for transmission electron-microscope (TEM) studies at an accelerating voltage of 200 kV. During the determination of the densities of radiation defects and

Table 1
Chemical compositions of pressure vessel materials

Steel type	wt%									
	Si	Mn	P	S	Cu	Ni	Cr	Mo	C	V
15Kh2MFA	0.27–0.37	0.39–0.48	0.011–0.016	0.012–0.018	0.12–0.14	0.19–0.27	2.52–3.00	0.64–0.71	0.13–0.18	0.25–0.31
SV-10KhMFT	0.15–0.35	0.97–1.03	0.029–0.036	0.012–0.013	0.15–0.21	0.09–0.29	1.37–1.58	0.43–0.50	0.05–0.07	0.19–0.23
15Kh2NMFA	0.17–0.37	0.3–0.6	<0.02	<0.02	<0.2	1.0–1.5	1.8–2.3	0.5–0.7	0.13–0.18	0.1–0.12
25Kh3NM	0.44	0.49	0.024	0.018	0.10	1.02	3.03	0.40	0.23	–

precipitates, the thickness of specimens was measured using the convergent beam electron diffraction method [12] giving an accuracy of at least 5%. The specimens for TEM studies were cut from halves of fractured Charpy specimens and were prepared by electrolytic polishing using a Struers electropolisher (Austria) with an electrolyte of 10% $HClO_4$ and 90% methanol at −70°C to −60°C just before placing them into the microscope.

The chemical compositions of grain boundaries were determined using AES [13] with a spectrometer Microlab Mk II (VG Scientific, England) at a beam energy of primary electrons of 5 keV. To obtain vacuum-pure surfaces, the fracture of specimens for AES studies was carried out directly in the vacuum chamber of the spectrometer at a pressure of approximately 10^{-10} mbar. The material for these fractures was obtained from the halves of tested Charpy specimens.

3. Results

Table 2 details typical results of fractographic studies applied to the RPVS in the initial state, irradiated in various conditions, or annealed and re-irradiated. The results permit the following conclusions to be drawn concerning grain boundary and intragranular phosphorus segregation and their evolution under irradiation and recovery annealing.

(1) Irradiation of the steels in conditions characteristic of RPV operation, results in significant changes in Charpy specimen fractures as compared with unirradiated specimens at comparable testing temperatures. For instance, in fractures of specimens tested in the range of US temperatures, in addition to the regions with ductile dimple fracture, the regions with ductile intergranular fracture are detected (Fig. 1(a)). Their proportion of the general area of the fracture surfaces reaches 10–15%. The appearance of such regions in the fractures is possible only when the grain boundaries are decorated with precipitates of phases in such a way that at the interface boundaries of these precipitates, phosphorus segregation (i.e., intragranular segregation) was proceeding as a consequence of thermal aging or irradiation.

(2) A reduction in testing temperature downwards to the DBTT induces an important decrease in the proportion of ductile and ductile intergranular fracture in general areas of fracture surfaces of irradiated specimens (Table 2).

Simultaneously, a number of regions appear, where transcrystalline fracture is registered that is formed from cleavage or quasi-cleavage (Table 2). It should be noted that in base metal, irradiation induces the appearance of an appreciable number of regions with brittle intergranular fracture (Fig. 1(b)). Their proportion in general areas of fracture surfaces usually constitutes 15–30% (Table 2). However, for some grades of steels it can

Table 2
Summary data of fractographic analysis results for Charpy specimens [10][a]

Material (wt%)	Specimen types	NPP	Fluence $\times 10^{23}$ (nm^{-2})	Condition	Test temperature (°C)	DBTT (T_k) (°C)	Absorb. energy (J)	Point on the curve KCV–T	Quota of different fracture modes (%)				
									Ductile	Quasi-cleavage	Cleavage	Inter-granular	Ductile intergranular
25Kh3NM P = 0.018 Cu = 0.10	BM Charpy	Cover from experimental PWR	–	Unirradiated, heat influence – 60 000 h	−150	−22	5.4	LS	–	35	–	65	–
					−35		46	DBT	10	5	5	65	15
					100		191	US	85	–	–	–	15
25Kh3NM P = 0.018 Cu = 0.10	BM Charpy	Trepan from experimental PWR	1.6	Irradiated	0	107	18	LS	10	15	10	65	–
					113		47	DBT	35	10	–	45	10
					200		108	US	90	–	–	–	10
25Kh3NM P = 0.018 Cu = 0.10	BM Charpy	Trepan from experimental PWR	6.5	Irradiated	23	168	5	LS	10	20	–	70	–
			3.7		175		40	DBT	40	5	5	40	10
			3.7		225		90	US	90	–	–	–	10
Sv-10KhMFT P = 0.0375 Cu = 0.18	WM subsize Charpy	Templet from WWER-440 Kozloduy NPP-2	6.7	Irradiated	−50	91	0.4	LS	10	75	10	5	n/d
					150		8.2	DBT	80	10	5	5	n/d
					250		9.6	US	100	–	–	–	n/d
Sv-10KhMFT P = 0.0375 Cu = 0.18	WM subsize Charpy	Templet from WWER-440 Kozloduy NPP-2	6.7	Irradiated + annealed 475°C – 150 h	−50	11	1.5	LS	15	75	5	5	n/d
					25		7.7	DBT	30	50	15	5	n/d
					250		14.4	US	100	–	–	–	n/d
Sv-10KhMFT P = 0.0375 Cu = 0.18	WM subsize Charpy	Templet from WWER-440 Kozloduy NPP-2	6.7	Irradiated + annealed 560°C – 2 h	−50	−24	2.9	LS	20	65	10	–	n/d
					−37		6.7	DBT	30	40	25	–	n/d
					150		15.2	US	90	5	–	–	n/d
Sv-10KhMFT P = 0.039 Cu = 0.17	WM Charpy	Surveillance samples	–	Unirradiated	−150	10	1.6	LS	–	65	35	–	–
					−12		41	DBT	35	40	25	–	–
					175		135	US	100	–	–	–	–
Sv-10KhMFT P = 0.039 Cu = 0.17	WM Charpy	Surveillance samples WWER-440	0.9	Irradiated	60	100	24.3	LS	30	60	10	–	–
					100		43.9	DBT	60	35	5	–	–
					160		83.1	US	90	–	–	–	10

Table 2 (Continued)

Material (wt%)	Specimen types	NPP	Fluence $\times 10^{23}$ (nm^{-2})	Condition	Test temperature (°C)	DBTT (T_k) (°C)	Absorb. energy (J)	Point on the curve KCV-T	Quota of different fracture modes (%)				
									Ductile	Quasi-cleavage	Cleavage	Intergranular	Ductile intergranular
Sv-10KhMFT P = 0.039 Cu = 0.17	WM Charpy	Surveillance samples WWER-440	0.9	Irradiated + annealed 470°C – 70 h	0	25	10.2	LS	10	55	35	–	–
					20		48.6	DBT	40	35	25	–	–
					100		137.2	US	85	5	5	–	5
Sv-10KhMFT P = 0.039 Cu = 0.17	WM Charpy	Surveillance samples WWER-440	0.9	Re-irradiated	−50	85	5	LS	–	65	35	–	–
					60		51	DBT	55	30	15	–	–
					250		95	US	90	–	–	–	10

[a] BM – base metal; WM – weld metal; LS – lower shelf; DBT – ductile to brittle transition region; US – upper shelf.

reach 70% (Table 2). The appearance of brittle intergranular fracture implies that grain boundary phosphorus segregation is occurring.

(3) A further reduction in Charpy test temperature downward to lower shelf values, results in practically complete disappearance of regions with ductile and ductile intergranular fracture in the fracture surfaces (Table 2). At the same time some reduction in the proportion of regions with intergranular fracture (in those materials where it occurred) in general areas of fracture surfaces is observed (Table 2); the proportion of cleavage and/or quasi-cleavage increases (Table 2).

(4) Several experimental data (Table 2) show that long-term aging of unirradiated RPVS at 270°C/60 000 h can also induce grain boundary phosphorus segregation. Moreover, the proportion of the regions with brittle intergranular fracture in Charpy specimens can reach 65% (Table 2). For Charpy of the same steel, irradiated to different neutron fluences (1.6×10^{23} and 4.8×10^{23} nm^{-2}) at 270°C, the proportion of regions with brittle intergranular fracture constitutes 65–70% as well (Table 2).

Thus, comparative fractographic investigations of a wide range of RPVS (base and weld metal) in initial and irradiated states have shown that the principal feature of fracture surfaces of Charpy specimens in the irradiated state (and in some cases after long-term aging) is the replacement of completely transcrystalline fracture to mixed types of fractures with brittle and/or ductile intergranular fracture. Moreover, it should be noted that for each of the grades of steels studied (i.e., base or weld metal) the temperature interval, where the grain boundary segregation occurs and also the proportion of brittle intergranular component in the fracture surface are higher, the higher is the concentration of phosphorus in the steel.

It should be emphasized that the area fraction of the regions with ductile intergranular fracture in fracture surfaces of any steel is not connected directly to the level of phosphorus segregation at interface boundaries of the corresponding precipitates. It is largely defined by the proportion of grain boundary surfaces decorated with such precipitates relative to the whole grain boundary surface.

The influence of phosphorus concentration on the proportion of brittle intergranular fracture is rather complicated. For instance, in specimens made from base metal, in which phosphorus content is significantly lower that in weld metal, the fraction of regions with brittle intergranular fracture in irradiated full-size Charpy specimens is much higher in comparable conditions. Apparently, this is caused by differences in microstructure of these materials. Based on the data given in [14], it would be reasonable to propose the following explanation of the observed propensity of full-size Charpy specimens made from irradiated base metal of Russian

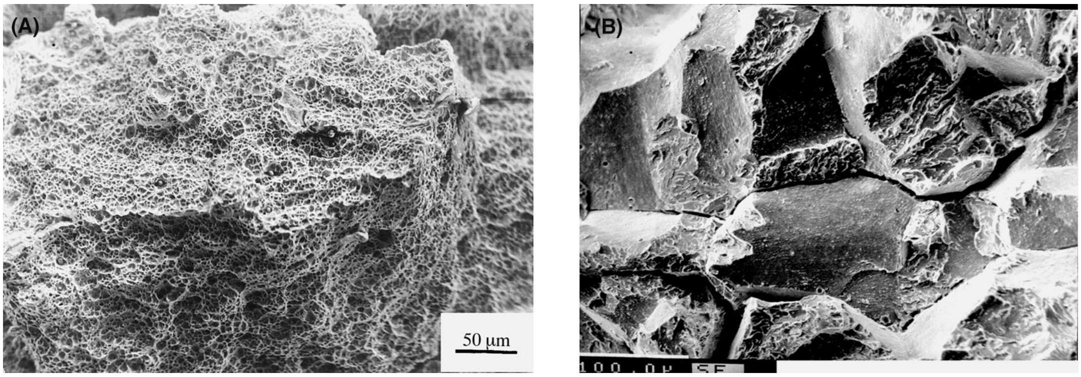

Fig. 1. Typical regions of Charpy specimen fractures with intergranular fracture: (a) ductile; (b) brittle.

grades to brittle intergranular fracture as compared with similar Charpy specimens made from irradiated weld metal of Russian grades. As metallographic researches show, the microstructure of base metal is homogeneous and consists of tempered bainite. Compared to base metal, the grains of weld metal consist not only of tempered bainite but at the locations of high-angle grain boundaries of the initial austenite grains, along with intergranular fracture in Charpy testing predominately occurs, grains of globular alpha-ferrite without carbide precipitates are also present [10]. Alloying elements Mo and Mn produce a sharp decrease in solubility of phosphorus in alpha-ferrite [14] and, accordingly, can result in a reduction of the level of phosphorus segregation along high-angle grain boundaries in weld metal. In favour of this hypothesis is the fact that in Charpy tests with full-size specimens made from base and weld metal of American grade steels that have the same homogeneous structure of tempered bainite over the sections of former austenite grains, the appearance of brittle intergranular fracture in Charpy specimens is equally likely (at high enough concentrations of phosphorus in steel) [10].

(5) Recovery annealing applied to irradiated RPVS results in significant changes of fractography at comparable temperatures of Charpy tests. However, the degree and character of these changes depend on the temperature of recovery annealing and subsequent rate of cooling. In cases where the temperature of recovery annealing is 450–475°C and the duration does not exceed 150 h, then at fast subsequent cooling an appreciable decrease in the proportion of the regions with ductile intergranular fracture relative to the total area of fracture surface occurs (Table 2). At the same time, an increase to the appropriate value occurs for the regions with ductile fracture.

It is essential that recovery annealing at 450–475°C does not result in reduction of the fraction of the regions with brittle intergranular fracture, but even induces its increase in some cases (Table 2).

Recovery annealings at 560°C result in further reduction in the fraction of the regions with ductile intergranular fracture (Table 2). Furthermore, they result in significant reduction in the fraction of the regions with brittle intergranular fracture (Table 2).

Table 3 lists the typical microstructural features found from TEM for RPVS in the initial state, irradiated in different conditions, and annealed and re-irradiated. The results provide a snapshot of the general aspects of radiation-induced structural changes induced by irradiation and also of the evolution of the structure resulting from recovery annealing.

1. The most important radiation-induced structural changes in RPVS are the formation of radiation defects and two types of precipitates: disc- and round-shaped (Table 3). Radiation defects in steels are observed as black dots and dislocation loops with a resolved line of 'zero' contrast [10]. The density of radiation defects is $\leqslant 10^{16}$ cm^{-3} at fast neutron fluences typical of WWER-440.
2. Irradiation induces a significant increase in the density of disc-shaped precipitates (vanadium carbides) (Table 3). These precipitates are 1–2 nm thick and have an average diameter $\geqslant 20$ nm in the initial state and $\approx$10 nm in the irradiated state (Fig. 2). The peculiar fact is that in numerous cases disc-shaped precipitates form chains along grain boundaries (Fig. 2(b)) in addition to formation of these precipitates in the volume of the grains (Fig. 2(a)). The density of disc-shaped precipitates under irradiation conditions typical of RPVS operation conditions can reach $(5–6) \times 10^{16}$ cm^{-3}. In the initial state their density is $\approx(0.5–0.8)\times10^{15}$ cm^{-3}.
3. Irradiation of RPVS also induces the formation of round-shaped precipitates (copper-enriched) of rather small sizes: $\approx$2–3 nm (Fig. 3, Table 3). These precipitates are distributed uniformly throughout the grains of the metal. Their density in the considered irradiation conditions can reach 2×10^{18} cm^{-3} (Table 3).

Table 3
Data on densities and average sizes of radiation defects, disk-shaped and rounded precipitates in weld metal before irradiation, after irradiation and after recovery annealing [10]

Condition	n_{loops} $\times 10^{15}$ (cm^{-3})	$\langle d \rangle_{\text{loops}}$ (nm)	n_{disk} $\times 10^{15}$ (cm^{-3})	$\langle d \rangle_{\text{disk}}$ (nm)	n_{rounded} $\times 10^{15}$ (cm^{-3})	$\langle d \rangle_{\text{rounded}}$ (nm)
Specimens from WWER-440 NPP Kozloduy-1 (P = 0.047%, Cu = 0.10%)						
Unirradiated quasi-archive	–	–	0.5–0.6	20.4	–	–
Re-irradiated $F = 0.5 \times 10^{23}$ m^{-2}	0.9–1.0	4–5	2.0–3.0	11.5	500–700	2.0–3.0
Re-irradiated + annealed 475°C, 150 h	–	–	1.0–1.5	11.5	20–30	3.0–4.0
Re-irradiated + annealed 560°C, 2 h	–	–	0.8–0.9	12.0	15–20	3.0–4.0
Surveillance samples from WWER-440 (P = 0.039%, Cu = 0.17%)						
Unirradiated	–	–	0.7–0.8	22.8	–	–
Irradiated $F = 0.9 \times 10^{23}$ m^{-2}	2–3	5–6	20–25	18.7	1700–2000	2.0–2.5
Irradiated + annealed 470°C, 70 h	–	–	2.5–3.0	25.6	40–50	3.0–4.0
Re-irradiated F $= 0.9 \times 10^{23}$ m^{-2}	1.5–2.0	5–6	10–15	23.3	400–500	3.5–4.5

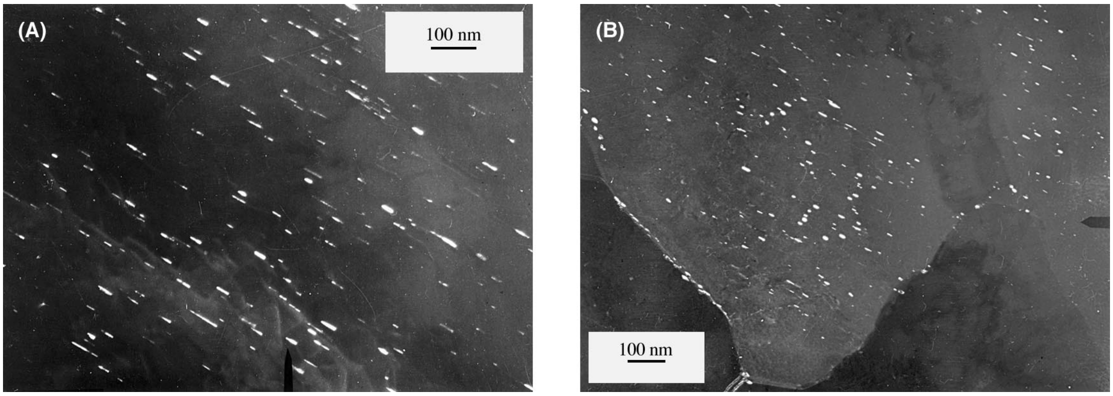

Fig. 2. Disc-shaped precipitates in irradiated RPVS: (a) inside grain body; (b) along grain boundaries.

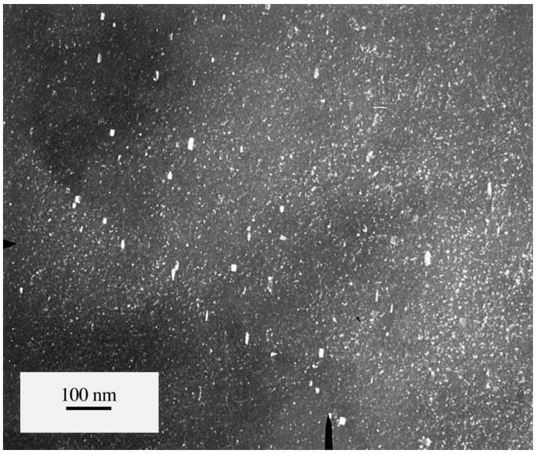

Fig. 3. Round-shaped precipitates in irradiated RPVS.

4. Recovery annealing at 470–475°C results in practically complete disappearance of radiation defects in steels (Table 3). At the same time the density of disc-shaped precipitates decreases more than one order of magnitude (Table 3). But a complete recover of the density of disc-shaped precipitates to the values typical of initial state does not occur. Recovery annealing at 470–475°C results in a significant (more than 25–40 times) decrease in the density of round-shaped precipitates and some increase in their average sizes (Table 3).
5. Recovery annealing at 560°C results in a stronger decrease in the density of disc-shaped precipitates (Table 3). However, complete recovery of their density (to the values typical of unirradiated steels) does not occur. Such annealing results in significant changes in size distribution of disc-shaped precipitates in comparison with annealing at 470–475°C. Thus, their average size can increase to values exceeding average sizes of disc-shaped precipitates in unirradiated steels (Table 3). Moreover, annealing at 560°C results in an even stronger decrease in the density of round-shaped precipitates compared with annealing at 470–475°C (Table 3).

The appearance of fractures in the regions with ductile intergranular fracture shows that the average distance between precipitates (at interface boundaries on which phosphorus segregation formed), which decorate the corresponding boundaries of matrix grains, should not exceed 1–5 μm. This value corresponds to the average distance between the centres of neighbour dimples at the regions with ductile intergranular fracture. TEM studies have shown that only disc- and round-shaped precipitates of high enough density satisfy this condition in irradiated weld metal. It should be noted here that decoration of matrix grain boundaries with disc-shaped precipitates is not necessarily observed. (Fig. 2(b)). As shown earlier, precipitates of this type arise mainly in grain interiors.

The AES method has been applied to examine a fracture surface in the region with brittle intergranular fracture of a specimen made from steel 25Kh3NM from the cover of a prototype reactor after operation for 60 000 h. The cover had undergone heat treatment (at 270–290°C) without irradiation. Fig. 4(a) shows a typical Auger electron spectrum obtained for this surface. The phosphorus peak is clearly seen in the spectrum and implies that the increase in phosphorus concentration at the grain boundary is a few orders of magnitude greater in comparison with its volume concentration in the matrix. The peaks of the other key elements in the steel composition are also seen.

Similar results have been obtained for irradiated RPVS 25Kh3NM of a prototype reactor following operation for 60 000 h. Fig. 4(b) shows the Auger spectrum obtained in the region with brittle intergranular fracture, confirming enrichment of grain boundaries with phosphorus. Fig. 4(c) shows the peak height ratio (PHR) for phosphorus and iron peaks vs the dose of radiation, where the peaks are measured at grain boundaries in the region of brittle intergranular fracture. This curve was measured through the thickness of the RPV wall [15,16]. It can be seen from Fig. 4(c) that with increase in fast neutron fluence the enrichment of grain boundaries with phosphorus increases thus confirming that an appreciable contribution to grain boundary phosphorus segregation arises from radiation-induced diffusion.

Thus, in the regions of fracture surface with intergranular fracture, grain boundary phosphorus segregation is observed (resulting from both irradiation and long-term aging).

In addition, the AES method has been applied to a specimen made from steel 15Kh2MFA irradiated to a neutron fluence 1.68×10^{23} nm^{-2} at the region of ductile intergranular fracture.

Fig. 5(a) illustrates the Auger electron spectrum taken from the surface in the region of ductile intergranular fracture shown in Fig. 5(b). The spectrum

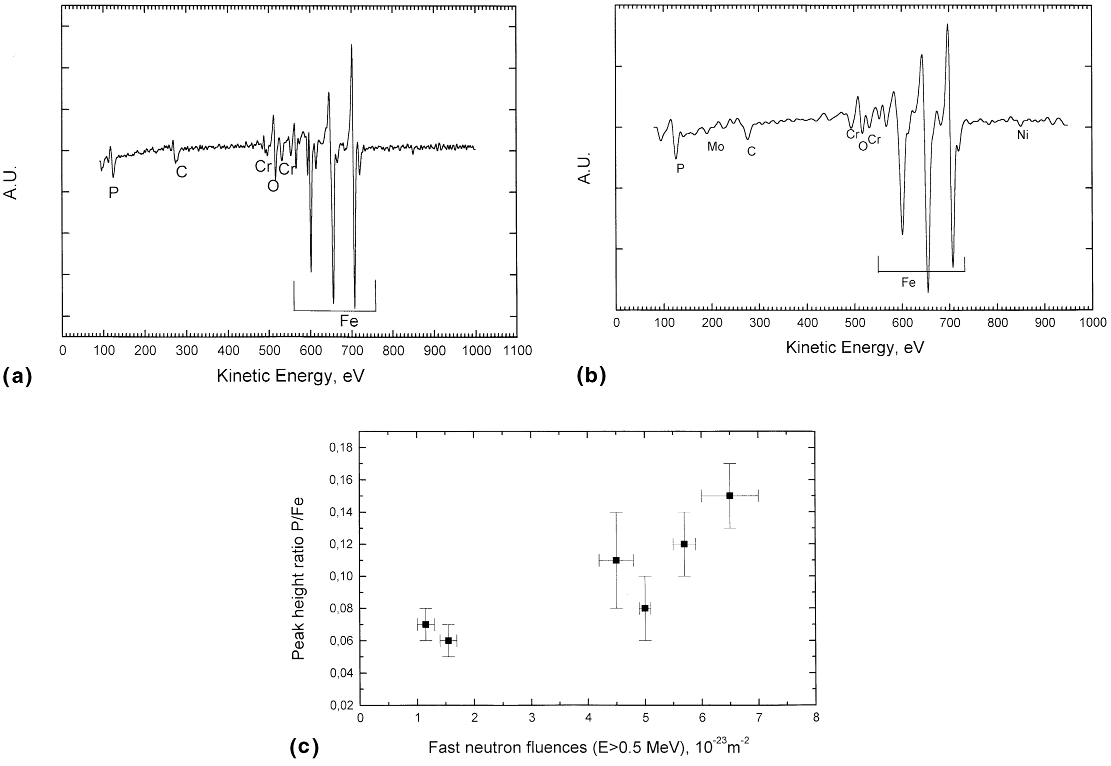

Fig. 4. Auger electron spectra of steel 25Kh3NM (base metal) of prototype reactor: (a) heating (270–290°C, 60000 h), unirradiated; (b) irradiated, $F = 6.5 \times 10^{23}$ nm^{-2}; (c) dose dependence of P/Fe PHR at grain boundaries in the regions of brittle intergranular fracture.

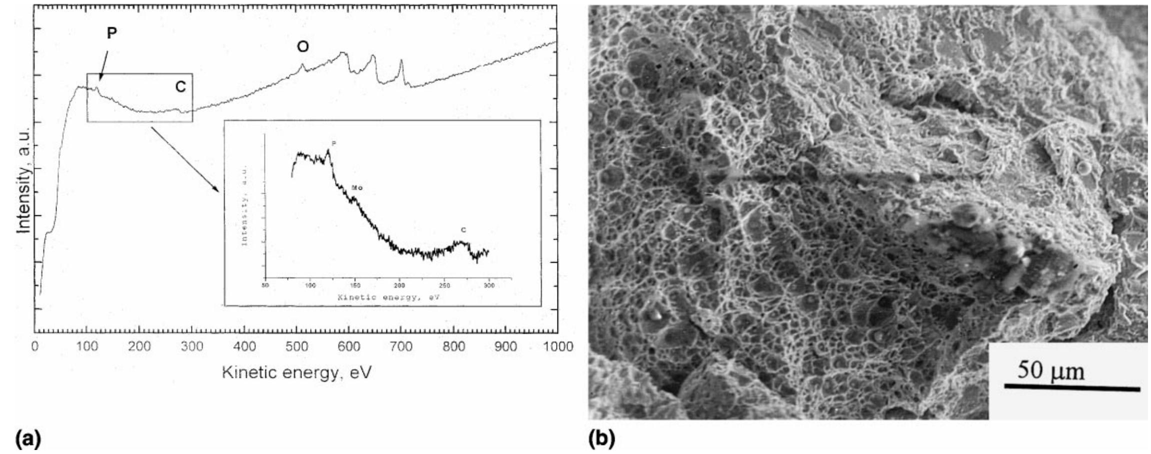

Fig. 5. Phosphorus segregation in the region of ductile intergranular fracture: (a) Integral Auger electron spectrum measured from fracture surface in (b); (b) the region from which the Auger electron spectrum presented in (a) has been measured.

contains Auger electron peaks of the elements Fe, P, Mo, C, O. Due to complexity of the geometry of the analysed fracture surface the spectrum is very 'noisy'. For this reason, even using the procedure of smoothing of the spectrum it was not possible to obtain a differential spectrum for quantitative evaluation. However, taking into account resolution, achievable for the AES method, it may be concluded that phosphorus is present on the surface of the ductile intergranular fracture at levels $\leqslant 5\%$. Therefore, phosphorus concentration in the region of ductile intergranular fracture is much higher than its volume concentration in the steel.

4. Discussion

The above experimental data allow the following conclusions to be made on the behaviour of Russian RPVS resulting from irradiation and recovery annealing.

As described earlier, irradiation induces the formation of radiation defects (dislocation loops), copper-enriched (round-shaped) precipitates and also increases the density of ultrafine dispersed vanadium carbides (disc-shaped precipitates), which are present in the steels in the initial state. Furthermore, irradiation induces phosphorus segregation of two types: grain boundary and intragranular (at interface boundaries of precipitate/matrix type).

Direct investigation of enrichment with phosphorus of fracture surfaces of brittle intergranular fracture confirms phosphorus segregation induced by irradiation.

The irradiation temperatures (250–290°C), at which grain boundary phosphorus segregation arises in RPVS, are much lower than characteristic temperatures for the development of classical temper embrittlement (400–500°C) [4]. However, practically in all experimental studies on temper embrittlement the periods of thermal aging did not exceed 10 000–20 000 h [5]. These time intervals approximately correspond to the RPV lifetime. It is known that an affinity between phosphorus and grain boundaries in steels at >600–650°C is not observed, and monotonically increases with decrease in temperature downward from these values [4]. At low temperatures the process of grain boundary phosphorus segregation is controlled exclusively by the kinetic factor, since the steady-state level of phosphorus in intergranular segregation increases monotonically with decrease in temperature (<600–650°C) [4]. Therefore, an increase in duration of thermal aging (at least one order of magnitude in comparison with the overall experimental database obtained on temper embrittlement) can change considerably the character of the temperature dependencies typical of intergranular phosphorus segregation just in the low-temperature region. The experimental data confirm the fact that long-term (60 000 h) aging of RPVS without irradiation at 270°C can result in intergranular phosphorus segregation (Table 2). Moreover, the proportion of brittle intergranular fracture can reach 65% (Table 2). The proportion of brittle intergranular fracture, enriched with phosphorus, for Charpy specimens made from the same steel irradiated in RPV to different neutron fluences (1.6×10^{23} and 4.8×10^{23} nm^{-2}) at 270°C is also 65–70% (Table 2). Firstly, these data (as well as the data on the level of grain boundary phosphorus segregation in these specimens obtained using AES) show that the dependence of the proportion of brittle intergranular component in fractures of Charpy specimen vs phosphorus concentration in grain boundary segregation demonstrates saturation. At the stage of saturation further increase in phosphorus concentration along grain boundaries caused by radiation-induced diffusion does not essentially affect the degree of brittle intergranular fracture in the fracture surfaces of irradiated Charpy specimens. Secondly, this situation can arise, if the process of radiation-induced diffusion of phosphorus and the process of competitive distribution of phosphorus take place such that partial segregation to interface boundaries of precipitates occurs instead of segregation only to high-angle boundaries of the initial austenitic grains. Due to these facts, the proportion of brittle intergranular fracture can, in principle, stay unchanged or even decrease. Such data are given in [30,31], where irradiated and re-irradiated RPVS with different concentrations of copper and phosphorus annealed to obtain a coarse-grain structure and temper embrittlement were examined. It is shown in [31] that the dependence of the proportion of brittle intergranular fracture on phosphorus concentration in grain boundary segregation reaches a stage of saturation. Thus, the change of grain boundary phosphorus concentration by approximately three times (within the interval 15–47%) achieved by temper embrittlement or irradiation does not affect, practically, the degree of brittle intergranular fracture. It is worth mentioning also that after annealing and irradiation in fractures of specimens made from steel with low copper concentration (0.01 wt%) and high phosphorus concentration (0.017 wt%), the degree of brittle intergranular fracture is higher than in steel with the same phosphorus concentration (0.017 wt%) and high copper concentration (0.16 wt%) [31]. The authors [31] ascertained that the phosphorus concentration at the boundaries of the initial austenitic grains in the steel with low and high copper concentration is approximately the same. Moreover no copper segregation was revealed (neither by the method FEGSTEM nor AES). Therefore, the synergism of copper and phosphorus influence can be explained only based on the existence of intragranular phosphorus segregation to interface boundaries of copper precipitates.

The available experimental data do not allow unambiguous resolution of the radiation component in grain boundary phosphorus segregation for RPVS irradiated at operating temperatures of 250–290°C, for 200 000–300 000 h (RPV lifetime).

An analysis of experimental data obtained in fractographic studies shows that the decrease in the proportion of brittle intergranular fracture on fracture surfaces of irradiated specimens and also specimens that had undergone temper embrittlement is observed at identical temperatures ($\geqslant 560$°C). This implies that the decay in grain boundary phosphorus segregation occurring during recovery annealing is independent of the mechanism of formation of this segregation: temper brittleness or radiation-induced diffusion. Moreover, the decay is independent of the relative contributions of these mechanisms. This situation looks reasonable, because the formation and decay of grain boundary phosphorus segregation in steels are reversible in temperature, and are described analytically using state (potential) functions [4]. Thus, the prehistory of such segregation does not affect the details of their decay occurring during recovery annealing. The above reasons explain why recovery annealing applied to RPVS irradiated at 450–500°C does not result in a decrease in the brittle intergranular component in the fracture surfaces of the specimens (Table 2). In contrast, annealing at indicated temperatures could result in an increase in the level of the segregation, if the duration is long enough. Despite this, the major fraction of recovery of radiation-induced DBTT shift results from recovery annealing at specified temperatures (Table 2). This strongly suggests that the contribution of grain boundary phosphorus segregation to RPVS embrittlement is insignificant. Such segregation is responsible for only a fraction of the DBTT shift, which does not recover during recovery annealing.

It should be noted that the most convincing direct experimental support of intragranular phosphorus segregation as a result of irradiation are the regions with ductile intergranular fracture observed on fracture surfaces of Charpy specimens (Table 2) and their enrichment with phosphorus as well. Obviously, when interface boundaries of some precipitates have affinity to phosphorus, then, all other conditions being the same, phosphorus segregation would form on them irrespective of their location inside a body or at the boundaries of matrix grains. It is worth mentioning here that in [7] the precipitates susceptible to phosphorus segregation at their interface boundaries during thermal aging were observed not only along boundaries, but also inside the bodies of matrix grains.

Ductile intergranular fracture is observed at temperatures of Charpy tests lying in the range of the US. This indicates that the tearing stress is low at the region of interface boundaries of these precipitates. In addition, it indicates that at the specified interface boundaries the level of tearing stress is lower than at those boundaries of the matrix grains where normal intergranular phosphorus segregation has appeared. This conclusion follows from the fact that at the temperatures of Charpy tests, lying in the range of the US, brittle intergranular fracture is not generally observed (Table 2).

The reasons for the distinction between the values of tearing stress at interface boundaries of precipitates and boundaries of matrix grains in the process of phosphorus segregation on them are not yet quite clear. They can be related to the characteristics of the structure of interface boundaries or to differences in the level of phosphorus segregation arising on them.

The appearance of the regions with ductile intergranular fracture in the fracture surfaces, by itself, indicates that tearing along interface boundaries of precipitates, decorating the boundaries of matrix grains, is realized at some lower level of the stress than along interface boundaries of the same precipitates located in the volume of matrix grains. One probable reason for this behaviour can be a partial superposition of the levels of segregation characteristic, apart, from the boundaries of matrix grains and interface boundaries of precipitates, decorating the boundaries of matrix grains.

Evolution of intragranular phosphorus segregation affected by annealing can proceed as dissolution of some fraction of precipitates with phosphorus segregation on their interface boundary (phosphorus segregation disappears completely in this case) or as partial dissolution of intragranular segregation. The second type of evolution can be a result of a relatively lower temperature interval of stability of intragranular phosphorus segregation at interface boundaries of some types of precipitates (in comparison with grain boundary segregation) [4].

It is also important to note that during annealing at 470–475°C the evolution of intragranular segregation can be rather complicated, since the temperature intervals of stability of intragranular phosphorus segregation for interface boundaries of different precipitates can differ. The data obtained using fractography (Table 2) show that in steel (similarly to grain boundary segregation) the precipitates with phosphorus segregation at their interface boundaries, which are stable at this annealing temperature, are present.

Data from the literature confirm that both grain boundary and intragranular phosphorus segregation can result in an increase in DBTT temperature [4,5,7].

Published research on reversible temper embrittlement in steels demonstrate that grain boundary segregation of impurities (primarily, phosphorus) does not induce a reduction in US level in transition curves (even if the shift of DBTT to positive temperatures is significant) [10,31].

In contrast, there is little literature data devoted to the influence of intragranular phosphorus segregation, but it appeared that their formation in steels induces a reduction in US level in transition curves [6,10]. These phenomena reveal essential differences between grain boundary and intragranular phosphorus segregation, which appear at precipitate/matrix interfaces.

The limited number of experimental data on phosphorus segregation to interface boundaries in steels does not allow an unambiguous answer as to why the absorption energy at the US decreases, if they appear. Such reduction of the absorption energy at US could be caused by a reduction in plasticity of steels due to phosphorus segregation at interface boundaries (at the rates of deformation characteristic of Charpy tests). A reduction in US could also be caused by a decrease in the effective section of specimens due to the presence of phosphorus segregation at precipitate/matrix interfaces. The latter can be a consequence of a local reduction in tearing stress in the region of precipitate/matrix interfaces, where phosphorus segregation has occurred. The above two reasons could be true in combination in the case considered.

The characteristics of phosphorus segregation to precipitate/matrix interfaces also permit an explanation for the few observations of an increase in the level of US to values exceeding initial level (i.e., US level in unirradiated steels) resulting from annealing of irradiated RPVS. One of the most probable reasons is the following. Thermal aging of RPVS in the initial state can induce phosphorus segregation to precipitate/matrix interfaces (for certain precipitates, which exist in the steel in the initial state and susceptible to phosphorus segregation). Two processes may proceed in the steel following irradiation: the appearance of new locations for intragranular phosphorus segregation and an increase in phosphorus concentration in existing segregant locations at interface boundaries. This would provide an extra reduction in US level and also DBTT shift. If the schedules of recovery annealing induce a decay of phosphorus segregation at interface boundaries of all precipitates that existed in the steel (or a decrease in phosphorus concentration in them downward to levels which are lower than initial) then the US level can increase upward to levels exceeding the initial (unirradiated state). In addition, it can induce recovery of DBTT to temperatures that are lower than the DBTT of the steels in the initial (unirradiated) state.

Numerous experimental data exist in the literature concerning the influence of the temperature of thermal aging on the rate of grain boundary phosphorus segregation in steels, and also on the temperature interval of its stability [17,19–21]. Studies on the process of phosphorus segregation to precipitate interface boundaries are however quite limited. Isolated experimental data suggest a reversible character for phosphorus segregation to interface boundaries [6,7]. Some experimental data show that the temperatures for decay of intragranular phosphorus segregation are somewhat lower than the temperatures of decay of grain boundary phosphorus segregation. The temperatures of decay of grain boundary phosphorus segregation are usually ~600–650°C [17–21]. On the other hand, in [8] it is shown that a complete recovery of DBTT and US levels in alloys of Fe–P irradiated at 50°C to a neutron fluence $\sim 10^{19}$ n cm^{-2}, resulting from recovery annealing, occurs at 420°C. There are enough convincing arguments in this paper confirming that DBTT and US level shifts in Fe–P alloys are caused by intragranular phosphorus segregation.

It should be emphasized that a complete recovery of US level for RPVS irradiated at 250–290°C is observed after annealing at ~420°C as well [23,28,29].

The temperatures of annealing of phosphorus segregation at precipitate interface boundaries (as distinct from grain boundary phosphorus segregation) can be determined by two processes:

1. Dissolution of phosphorus, which forms segregation at interface boundaries, and its migration into the matrix.
2. Dissolution of precipitates, at interface boundaries of which phosphorus segregation existed, and disappearance of the corresponding segregation.

Minimum values of annealing temperature necessary for the disappearance of phosphorus segregation at interface boundaries are defined by minimum temperatures at which at least one of the two above processes is realised. This property essentially distinguishes phosphorus segregation to interface boundaries from grain boundary phosphorus segregation. In the latter case, a disappearance of phosphorus segregation resulting from annealing (at <700°C) is possible only through the mechanism of its dissolution into the matrix.

Since the formation and behaviour of grain boundary phosphorus segregation in steels due to thermal aging can be described by means of potential or state functions, the phosphorus concentration in grain boundary segregation is independent of prehistory of the material (i.e., keeping steels at any temperatures <700°C). This means that the phosphorus concentration in grain boundary segregation after long-term annealing of irradiated RPVS depends only on annealing temperature (and can depend also on the rate of cooling after annealing).

A rather limited body of experimental data concerning the formation and behaviour of phosphorus segregation to interface boundaries resulting from thermal aging applied to steels is available at the moment [6,7]. These data indicate that the formation and behaviour of phosphorus segregation to interface boundaries induced by thermal aging can be described by means of potential functions as well. It can be assumed that phosphorus concentration in segregation at interface boundaries depends only on annealing temperature for long-term annealing.

The temperatures of the most intensive formation or decay of phosphorus segregation at interface boundaries can depend on the type of corresponding precipitates. The data from [7,8] specify that the temperatures of the most intensive formation of intragranular phosphorus segregation in RPVS investigated after thermal aging, apparently, are a little bit lower than the corresponding temperatures for grain boundary phosphorus segregation. However, this difference is not so significant to suppose dissolution of phosphorus segregation at interface boundaries in the matrix for annealing temperatures of 400–420°C.

As pointed out above, the disappearance of phosphorus segregation at interface boundaries (or on surfaces of radiation defects) induced by annealing (as distinct from grain boundary phosphorus segregation) can also occur due to dissolution of the corresponding precipitates (or radiation defects) in the matrix. The data on electron microscopy given in Table 3 show that annealing at 475°C results in the complete disappearance of radiation defects (dislocation loops) and a reduction in the density of round- and disc-shaped precipitates of approximately 35–40 and ~14 times, respectively. Annealing at 560°C reduces the density of these precipitates to a greater degree (but not up to initial values). These data show that in Russian grade RPVS, the disappearance of phosphorus segregation at interface boundaries induced by recovery annealing can be attributed to the mechanism of dissolution of the corresponding precipitates (and annealing of radiation defects) in the matrix.

The available experimental data indicate that in steels of American grade, annealing at ~470°C also induces dissolution of an appreciable fraction of the radiation-induced precipitates enriched with copper [22]. It could be expected that if phosphorus segregation on surfaces of these precipitates exists, then annealing, affecting dissolution of the precipitates in the matrix would result in disappearance of phosphorus segregation at the locations of interface boundaries of dissolved precipitates.

The results and considerations stated above lead to the conclusion that minimum annealing temperatures inducing disappearance of phosphorus segregation at interface boundaries for irradiated RPVS can be defined by the temperatures of dissolution of the corresponding precipitates in the matrix instead of the temperatures of dissolution of phosphorus segregation itself.

The following correlation is also of interest: practically for all irradiated RPVS given annealing at temperatures of 400–420°C complete recovery of the US level is achieved, but the degree of DBTT recovery varies between ~50% and ~80% [23–28]. This means that a fraction of the DBTT shift (RE) in RPVS, which is related to a reduction in US level, apparently, is responsible for the major fraction of RE in RPVS. Earlier in [10] experimental data were published, which indicated that among three mechanisms of embrittlement (potentially capable of providing a contribution to RE in RPVS) only two can cause a decrease in US level in transition curves accompanied with a DBTT shift, namely hardening and intragranular phosphorus segregation. Here the greatest contribution to RE proceeding in Russian RPVS with high phosphorus concentration arises from intragranular phosphorus segregation. The third mechanism, i.e., grain boundary phosphorus segregation, does not result in a reduction of US level. The data from [10] imply that for RPVS the contribution of grain boundary phosphorus segregation to the radiation-induced shift of DBTT (irradiation to neutron fluences $<10^{20}$ n cm^{-2} at ~250–300°C) does not exceed ~10–20%.

5. Conclusions

1. The principal feature of the fractography of irradiated (or in some cases long-term aged) Charpy specimens of RPVS is the transition from purely transcrystalline to mixed mode fracture, characterised by the presence of brittle and/or ductile intergranular fracture.
2. The presence of grain boundary phosphorus segregation in the fracture surfaces of RPVS specimens with intergranular fracture both following irradiation and long-term aging (~60 000 h) at operating temperatures of 270–290°C has been established.
3. The role of radiation-induced diffusion of phosphorus in the process of grain boundary phosphorus segregation has been established. Moreover, the existence of saturation in the dependence on the degree of brittle intergranular fracture vs phosphorus concentration in grain boundary segregation has been observed. This saturation can be one reason for the relatively low contribution of grain boundary phosphorus segregation to RE, as observed in RPVS.
4. The existence of intragranular phosphorus segregation, which provides an essential contribution to RE of RPVS, has been established experimentally for the first time. In addition, it has been demonstrated that the formation at interface boundaries of radiation-induced precipitates, decorating high-angle boundaries of the initial austenitic grains, results in the appearance of ductile intergranular fracture in the fracture surfaces of irradiated Charpy specimens.
5. It has been established that recovery annealing at 470–475°C reduces the amount of ductile intergranular fracture in fracture surfaces of irradiated Charpy specimens, whereas the level of brittle intergranular fracture either remains unchanged or increases. This means that annealing within the relevant temperature interval induces a component of intragranular phosphorus segregation but does not induce dissolution of grain boundary phosphorus segregation.
6. It has been demonstrated that recovery annealing at $\leqslant 560$°C (if the period of the treatment and the rate of cooling from the annealing temperature are sufficient) induces dissolution of both grain boundary and intragranular phosphorus segregation. The consequence is a disappearance of both brittle and ductile intergranular components in the fractures of Charpy specimens.

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