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Simulation of irradiation effects in light water reactor vessel steels – experimental validation of RPV-1

Stéphanie Jumel *, Jean-Claude Van Duysen

EDF/R&D site des Renardières, Route de Sens, 77818 Moret sur Loing, France
Laboratoire de Physique et Génie des Matériaux (LMPGM), Université des Sciences et Technologies de Lille I, 56655 Villeneuve d'Ascq, France

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Abstract

The REVE project (REactor for Virtual Experiments) was an international effort aimed at developing tools to simulate irradiation effects in materials of Light Water Reactors. In the framework of this project, a European team developed a first tool, called RPV-1, to simulate irradiation effects in light water reactor pressure vessel steels. This article is the fourth of a series dedicated to the presentation of RPV-1. It has a twofold objective:

– to show quantitative comparisons between experimental and RPV-1's simulation results;
– to demonstrate that RPV-1 can already be used to complement experimental programs.

To this end, RPV-1 has been used to reproduce the French experimental program ESTEREL which was aimed at quantifying the neutron spectrum effect between the surveillance capsules and the vessels of the French reactors, and to determine the best irradiation parameter to assess the behaviour of these vessels from the results of the surveillance program.

1. Introduction

Many key components in commercial nuclear reactors are subject to neutron irradiation which modifies their mechanical properties. So far, the prediction of the in-service behavior and the lifetime of these components have required irradiating materials in so-called 'experimental test reactors'.

A proactive way to complement this experimental approach is to develop physically-based computer tools to numerically simulate irradiation effects. The development of such tools, also called virtual test reactors (VTRs), started in the framework of the REVE Project (REactor for Virtual Experiments). This project (e.g. [1–3]) was a joint effort between Europe, the United States and Japan aimed at building VTRs able to simulate irradiation effects in pressure vessels and internal structures of light water reactors (LWRs). In this framework, the European team has built a first VTR, called RPV-1, designed for reactor pressure vessel steels (RPV steels) [3].

* Corresponding author.
E-mail addresses: stephanie.jumel@edf.fr (S. Jumel), jean-claude.van-duysen@edf.fr (J.-C. Van Duysen).

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RPV-1 relies on many simplifications and approximations and has to be considered as a prototype developed to clear the way. Long-term efforts will be required to complete it and to build successive generations of more and more sophisticated versions. Nevertheless, RPV-1 can already be used to complement experimental irradiation programs (understanding of results, assessment of material and irradiation conditions effects...). Its input and output data are similar to those of experimental irradiation programs carried out to assess the in-service behavior of reactor pressure vessels (for input data: neutron spectrum; irradiation temperature; Cu, Ni, Mn contents; grain size; dislocation density; tensile test temperature – for output data: irradiation-induced increase of yield stress; description of the irradiation-induced damage).

This article is the fourth of a series aimed at presenting RPV-1 [4–6]. It has a twofold objective:

- to show quantitative comparisons between experimental results and RPV-1's simulation results. These comparisons concern only the irradiation-induced evolution of yield stress. Quantitative comparisons about the irradiation-induced evolution of the microstructure will be given in a subsequent article.
- to demonstrate that RPV-1 can already be used to complement experimental programs. To this end, RPV-1 has been used to reproduce the French experimental program ESTEREL aimed at studying the neutron spectrum effect between the surveillance capsules and the vessels of the French reactors.

It should be mentioned that the simulations were carried out with standard parametrizations (mostly proposed by developers) of the codes used in RPV-1 [3–9]. The optimisation of these parametrizations is now in progress from large experimental databases and will be presented in subsequent papers.

2. Comparison of experimental and simulation results

A large program was carried out to compare experimental results and RPV-1's results obtained on pure iron, binary Fe–Cu alloys and RPV steels. Only some examples are given in this paragraph, they were obtained with the version 1.2.g of RPV-1. Section 2.3 presents additional results concerning RPV steels.

2.1. Pure iron

The conditions used to assess the quantitative character of RPV-1 on pure iron are summarized in Table 1. Neutron spectra representative of irradiation channels of the two experimental test reactors HFIR (USA) [10–12] and OSIRIS (France) [13] were used. The simulation results and available experimental results are compared in Figs. 1 and 2. In all cases, simulation results fall in a range of about ±20 MPa of experimental ones, which is

Table 1
Conditions used to assess the quantitative character of RPV-1 on pure iron

Reactor	Flux ($E > 1$ MeV) (10^{16} nm^{-2} s^{-1})	Irradiation temperature (°C)	Ref.
HFIR	7×10^2	≈60 150 300	[10–12]
OSIRIS	4.7	288	[13]

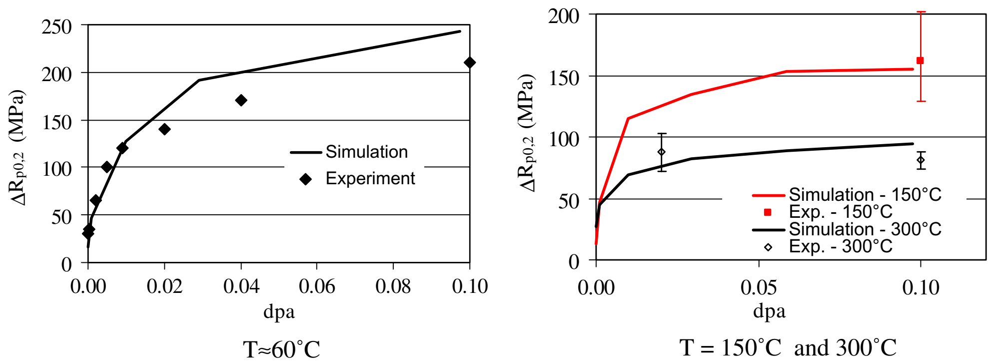

Fig. 1. Simulated evolution of the yield stress of pure iron irradiated in a channel of HFIR [Flux ($E > 1$ MeV) $= 7 \times 10^{18}$ nm^{-2} s^{-1}]. Comparison with experimental results [10–12].

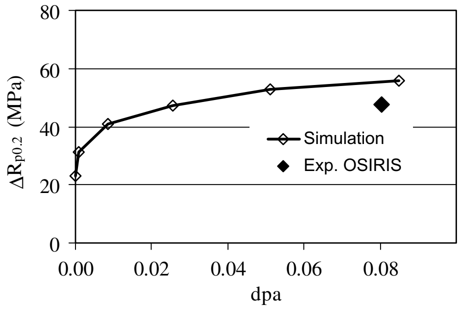

Fig. 2. Simulated evolution of the yield stress of pure iron irradiated at 288 °C in a channel of OSIRIS [Flux ($E > 1$ MeV) $= 4.7 \times 10^{16}$ nm^{-2} s^{-1}]. Comparison with experimental results [13] deduced from hardness measurements using the expression: $\Delta R_{P0.2} \approx 2.5\ \Delta\mathrm{Hv}$ [3,6].

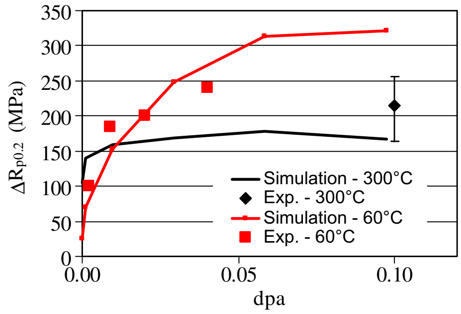

Fig. 3. Simulated evolution of the yield stress of an Fe–0.3%Cu model alloy, irradiated at 60 and 300 °C in a channel of HFIR [Flux ($E > 1$ MeV) $= 7 \times 10^{18}$ nm^{-2} s^{-1}]. Comparison with experimental results [12].

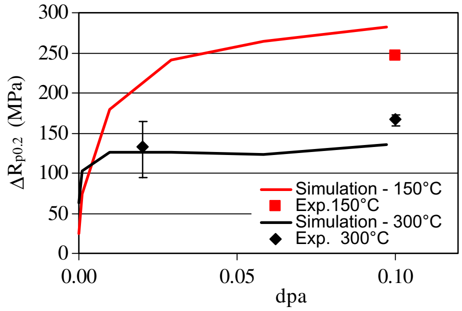

Fig. 4. Simulated evolution of the yield stress of an Fe–0.1%Cu model alloy, irradiated at 150 °C and 300 °C in a channel of HFIR [Flux ($E > 1$ MeV) $= 7 \times 10^{18}$ nm^{-2} s^{-1}]. Comparison with experimental results [14].

rather satisfactory (±20 MPa can be considered as the statistical dispersion of experimental irradiation-induced increases of yield stress on steels).

2.2. Fe–Cu model alloys

The quantitative character of RPV-1 on model Fe–Cu alloys was assessed with a neutron spectrum representative of one irradiation channel of the experimental test reactor HFIR [14,12]; the irradiation conditions are summarized in Table 2.

The simulation and experimental results are compared in Figs. 3 and 4. In all cases, the simulation results are in agreement with experimental ones and fall within the usual statistical dispersion of experimental irradiation programs: $\Delta\sigma \approx \pm 20$ MPa.

2.3. RPV steels

The quantitative character of RPV-1 on RPV steels was assessed with neutron spectra representative of the irradiation channels of two experimental reactors: HFIR [12,17] and HERALD [15]; the conditions are summarized in Table 3.

Table 2
Conditions used to assess the quantitative character of RPV-1 on Fe–Cu alloys

	Cu content (%)	Irradiation temperature (°C)	Ref
HFIR	0.1	150	[14]
Flux		300	
($E > 1$ MeV) =			
7×10^{18} nm^{-2} s^{-1}			
	0.3	60	[12]
		300	

The simulated and available experimental results are compared in Figs. 5 and 6. In all cases simulation results are in agreement with experimental ones and fall within the usual statistical dispersion of experimental irradiation programs: $\Delta\sigma \approx \pm 20$ MPa.

Other examples of validation on RPV steels are given in Section 2.3.

3. A simulated version of the French experimental program ESTEREL

As mentioned in [5], RPV-1 is still suffering from many weaknesses [3–6]: poor parameterization of the MFVISC Rate Theory code, weak sensitivity to Mn and Ni contents, rough approximation of the pinning forces, etc. However, it may already be used to complement experimental irradiation programs. This paragraph is aimed at demonstrating this capability by showing that RPV-1 allows to reproduce the French experimental program ESTEREL.

Table 3
Conditions used to assess the quantitative character of RPV-1 on RPV steels

Reactor	Steel			Flux ($E > 1$ MeV) 10^{16} nm^{-2} s^{-1}	Irradiation temperature (°C)	Ref.
	Cu (%)	Mn (%)	Ni (%)			
HFIR	0.14	1.30	0.87	7×10^2	55	[12]
HERALD	0.22	1.45	0.22	5	225	[15]

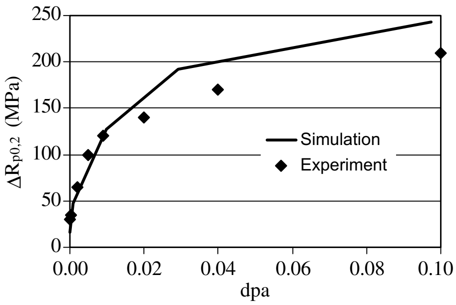

Fig. 5. Simulated evolution of the yield stress of a RPV steel irradiated at 55 °C in a channel of HFIR [Flux ($E > 1$ MeV) = 7×10^{18} nm^{-2} s^{-1}], Cu = 0.14%, Mn = 1.30%, Ni = 0.87%. Comparison with experimental results [12].

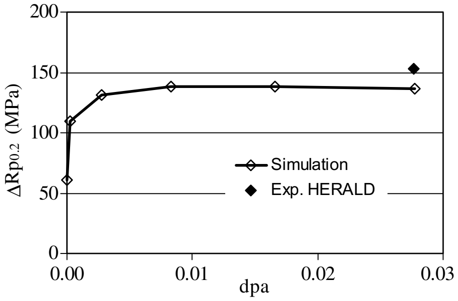

Fig. 6. Simulated evolution of the yield stress of a RPV steels irradiated at 225 °C in a channel of HERALD [Flux ($E >$ 1 MeV) = 5×10^{16} nm^{-2} s^{-1}]. Cu = 0.22%, Mn = 1.45%, Ni = 0.22%. Comparison with experimental results [15].

The experimental version and simulated version of ESTEREL are successively presented hereafter. The simulations were carried out with the version 1.3.c of RPV-1.

3.1. The experimental program

The ESTEREL program was carried out between 1989 and 1995 by the Commissariat à l'Energie Atomique (CEA) and Electricité de France (EDF) [16]. Its objective were

– to quantify the neutron spectrum effect between the surveillance capsules and the pressure vessels of the French reactors;
– to identify the most appropriate irradiation exposure parameter ($\phi_{E>1\text{MeV}}$, $\phi_{0.1\text{MeV}}$, ϕ_{dpa}) to assess the behaviour of these vessels from the results of the surveillance program.

The experimental conditions had to be as close as possible to the in-service conditions, so as to ensure that the results are as representative as possible. In order to meet these objectives, two low-alloyed Mn Ni Mo welds were specifically manufactured, two test reactors were used to carry out the irradiations, special devices (IRMA rigs,...) were developed and all experimental parameters were controlled with an extreme care (e.g. irradiation temperature: 288 ± 5 °C). The ESTEREL program was a large technical and financial effort. We reproduced it with RPV-1.

3.1.1. Irradiated materials

The two welds, named M1 and M2, were prepared with a manufacturing process (welding conditions, heat treatments,...) similar to that undergone by the French pressure vessel welds. Welds were preferred to base metals for their better metallurgical and mechanical homogeneities. The chemical compositions of M1 and M2 are given in Table 4. The two materials are low copper steels and mainly differ from their copper content: Cu = 0.048% for M1 and Cu = 0.095% for M2. Their irradiation-responses were studied in terms of:

– Charpy Ductile Brittle Temperature Transition (DBTT) shift. For each condition (material and reactor), transition curves were determined with 30 specimens; 15 of them were broken in the transition zone so as to determine precisely the DBTT;
– Yield stress increase. For each condition (material and reactor), 2 specimens were tested at −80 °C, −40 °C, 20 °C, 100 °C, 290 °C. Only the results obtained at 20 °C will be used in the following sections.

Table 4
Chemical compositions of materials M1 and M2 (wt%)

	C	Si	Mn	P	S	Ni	Cr	Mo	Co	Cu	V
RCC-M ed. 95	<0.100	0.15–0.60	0.80–1.80	<0.010	<0.025	<1.20	<0.30	0.35–0.65	<0.03	<0.07	<0.02
M1	0.073	0.43	1.59	0.008	0.008	0.76	0.16	0.58	0.010	0.048	0.003
M2	0.058	0.49	1.48	0.020	0.009	0.68	0.04	0.54	0.017	0.095	0.001

3.1.2. Irradiation conditions

Two experimental French reactors were used to carry out the ESTEREL program:

- the OSIRIS reactor (CEA-Saclay), using an irradiation channel with a neutron spectrum representative of that on the inner side of the French vessel;
- the SILOE reactor (CEA-Grenoble), using an irradiation channel equipped with a steel shield aimed at 'distorting' the neutron spectrum to get: (i) a neutron spectrum representative of that in the French surveillance capsules and (ii) a dpa rate similar that of the OSIRIS spectrum.

The spectra in both locations are shown in Fig. 7 where they are compared to those of the French power reactors. Spectra on the vessel and in OSIRIS, on the one hand, as well as spectra in the surveillance capsules and in SILOE, on the other hand, have very similar shapes. For neutrons with energy lower then 1 MeV, the OSIRIS and SILOE spectra accentuate the difference between the spectra in the capsules and on the vessel. The irradiations of the rigs lasted 185 and 187 effective full power days (EFPD) in SILOE and OSIRIS, respectively. It was expected that these time spans and the stacks of samples in the irradiation rigs would allow each weld to be irradiated with exactly the same number of dpa in both reactors.

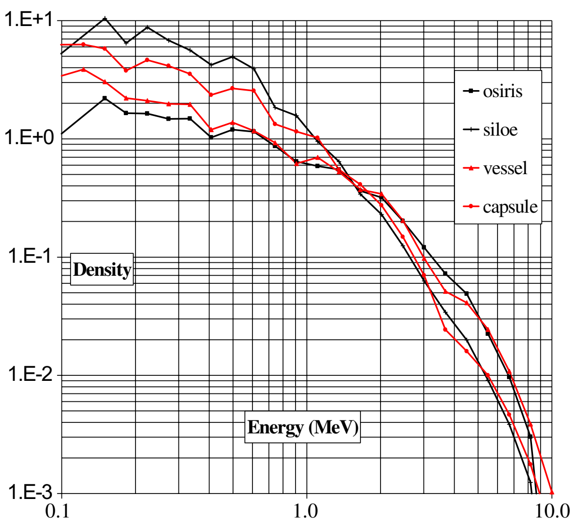

Fig. 7. Normalized neutron spectra in the surveillance capsules and on the inner side of the French pressure vessels as well as in the irradiation channels of OSIRIS and SILOE used in the ESTEREL program [16].

3.1.3. Results of the ESTEREL program

Due to the slight difference of copper content between M1 and M2 and the slight difference between the two spectra, very small discrepancies between the measured irradiation-responses were expected. The challenge of the ESTEREL program was to reveal and to quantify them.

Table 5 gives the DBTT shifts [measured at 56 J (ΔTK7) and at 0.9 mm of lateral expansion (ΔT0.9)] and the yield stress increases of M1 and M2 after irradiation in each reactor, versus several exposure parameters. It can be noticed that:

- in spite of the extreme care in the control of the irradiation conditions, the fluence $E > 1$ MeV and the number of dpa received by each weld is about 18% higher in OSIRIS than in SILOE;
- for each condition (material and reactor), the two methods of measurement of the DBTT shift lead to similar results;
- the embrittlement and hardening of each weld is higher after the OSIRIS irradiation than after the SILOE one.

For both materials, the higher embrittlement obtained in OSIRIS than in SILOE complies with the difference of fluence $E > 1$ MeV ($\phi_{E>1\mathrm{MeV}}$) and number of dpa (ϕ_{dpa}) obtained in both reactors. However, the fluence $E > 0.1$ MeV ($\phi_{E>0.1\mathrm{MeV}}$) is 20% lower for OSIRIS than for SILOE, which indicates that this fluence is totally inadequate to assess the behaviour of pressure vessels from the surveillance program.

3.1.4. Analysis of the experimental results

As mentioned in the previous section, $\phi_{E>0.1\mathrm{MeV}}$ could be easily discarded but a more refined analysis

Table 5
Increase of yield stress and DBTT shifts (at 56 J and 0.9 mm of lateral expansion) of materials M1 and M2, versus irradiation exposure parameters in OSIRIS and SILOE [16]

Weld	Reactor	$\Delta R_{P0.2}$ (MPa)	ΔTK7 (°C)	ΔT0.9 (°C)	mdpa	$\phi_{E>1MeV}$ (10^{23} nm^{-2})	$\phi_{E>0.1MeV}$ (10^{23} nm^{-2})
M1	SILOE	79.0	45.0	49.0	70.0	3.51	18.20
	OSIRIS	86.0	54.0	59.0	87.0	5.98	14.50
	Difference OSIRIS-SILOE	+7	+9	+10	+17	+2.47	−3.70
M2	SILOE	110.0	59.1	71.3	81.5	4.09	21.20
	OSIRIS	130.0	72.7	87.6	100.0	6.86	16.70
	Difference OSIRIS-SILOE	+20	+13.6	+16.3	+18.5	+2.77	−4.5

TK7: Ductile brittle transition temperature measured at 57 J.
ΔT0.9: Ductile brittle transition temperature measured at 0.9 mm of lateral expansion.

was needed to determine whether $\phi_{E>1MeV}$ or ϕ_{dpa} is the most appropriate exposure parameter.

The initial aim of the project was a direct comparison of the embrittlement levels induced by the irradiations in OSIRIS and SILOE, at the same number of dpa. As this number was about 18% lower in the SILOE irradiation, this direct comparison was not possible. To overcome this difficulty, the embrittlement levels produced in SILOE were extrapolated at the same number of dpa as in OSIRIS (from 70 to 87 mdpa for M1 and from 81.5 to 100 mdpa for M2) using the expression $\Delta DBTT_{Average} = K(\phi_{dpa})^{0.4}$, where $\Delta DBTT_{Average}$ is the average of the ΔDBTTs determined at 56 joules and at 0.9 mm of lateral expansion, K is a material-dependant constant. Similar derivation was made to compare the embrittlement levels at the same $\phi_{E>1MeV}$. Results are given in Table 6. For both materials, it can be noticed that the same ϕ_{dpa} leads to a slightly larger embrittlement in OSIRIS than in SILOE, while $\phi_{E>1MeV}$ gives very similar results. This suggests that $\phi_{E>1MeV}$ is the best exposure indicator to assess the behaviour of the pressure vessels from the results of their surveillance program. In all the cases, the spectrum effect between both locations is rather low (some degrees on the ΔDBTT).

3.2. A simulated version of the ESTEREL program

As for the experimental program, the challenge was to reveal and to quantify the very small discrepancies between the irradiation-responses of M1 and M2 in both reactors and to interpret them. This section gives the conditions of the simulations and the obtained results.

3.2.1. Simulations carried out

RPV-1 was run using the OSIRIS and SILOE neutron spectra (Fig. 7) and the chemical compositions of M1 and M2. The simulations were carried at out 288 °C with a dose up to 110 mdpa and dose

Table 7
Main parameters used in RPV-1

Name	Nature	Value
E_v^m	Vacancy migration energy (eV)	1.3
E_i^m	Interstitial migration energy (eV)	0.3
E_v^f	Vacancy formation energy (eV)	1.9
E_i^f	Interstitial formation energy (eV)	5
D_v^0	Vacancy diffusion pre-exponential factor	1
D_i^0	Interstitial diffusion pre-exponential factor	4×10^{-4}
E_bv	Binding energy of di-vacancies (eV)	0.2
E_bi	Binding energy of di-interstitials (eV)	1
ρ	Dislocation density (m^{-2})	10^{14}

Table 6
Comparison between the ΔDBTT induced by the OSIRIS irradiation and the ΔDBTT extrapolated at the same dose and fluence as in OSIRIS from the results of the SILOE irradiation [16]

Materials	OSIRIS $\Delta DBTT^{a}_{Average}$ ($\phi_{dpa}/\phi_{E>1MeV}$)	SILOE $\Delta DBTT^{a}_{Average}$ extrapolated to the same ϕ_{dpa} and $\phi_{E>1MeV}$ as in OSIRIS	
		Same ϕ_{dpa}	Same $\phi_{E>1MeV}$
M1	56.5 °C (87 mdpa/5.98 × 10^{23} nm^{-2})	51.3 °C	58.2 °C
M2	80.2 °C (100 mdpa/6.86 × 10^{23} nm^{-2} [a])	70.8 °C	80.2 °C

[a] average of the ΔDBTTs determined at 56 J and at 0.9 mm of lateral expansion.

rates similar to those of the experimental program: 6.81×10^{-9} dpa s^{-1} for OSIRIS reactor and 4.81×10^{-9} dpa s^{-1} for SILOE reactor. Parameters used in RPV-1 are given in Table 7.

3.2.2. Results of the simulations

The simulated increases of yield stress of both materials after irradiation are reported in Fig. 8 (versus the number of dpa), where they are compared to the increases of yield stress measured at 20 °C and calculated from the DBTT shifts with the expression $\Delta DBTT = 0.63\,\Delta R_{P0.2}$ [17]. All these results are summarized in Fig. 9. It can be noticed that:

– In spite of the small difference of copper contents between both materials, RPV-1 correctly repro-

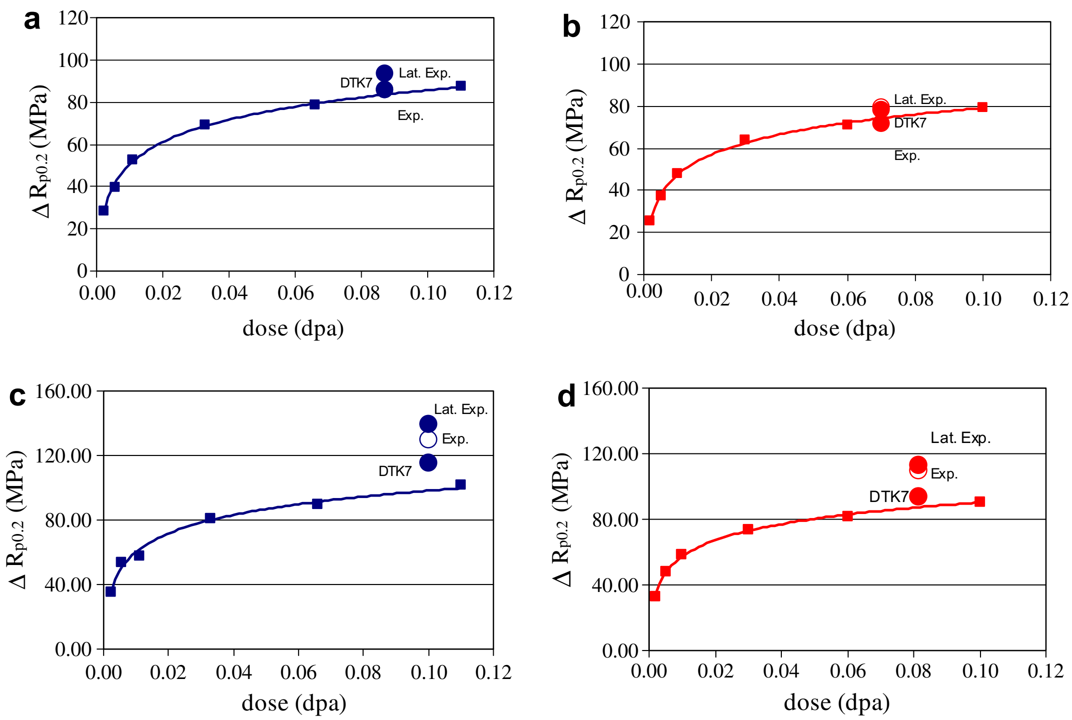

Fig. 8. Comparison of simulation (squares and curves) and experimental (circles) results. Lat. Exp.: increase of yield stress deduced from the DBBT shift measured at 0.9 mm of lateral expansion. DTK7: increase of yield stress deduced from the DBBT shift measured at 56 J. Exp.: increase of yield stress measured by tensile test. (a) Material M1 irradiated in OSIRIS. (b) SILOE. (c) Material M2 irradiated in OSIRIS. (d) SILOE.

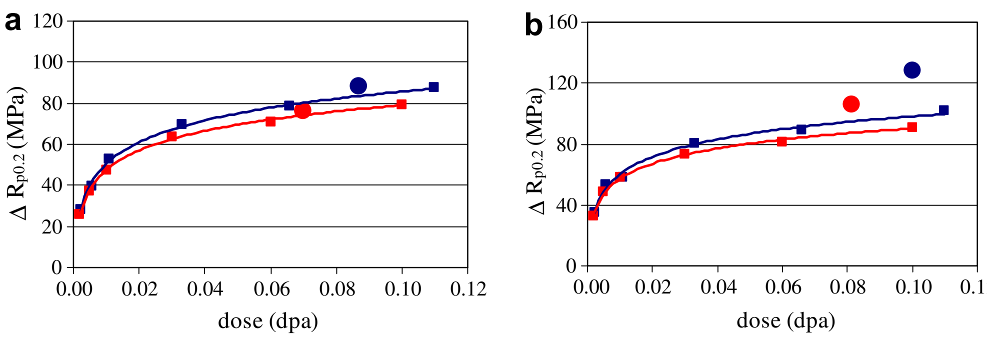

Fig. 9. Summary of the comparison between the simulation (squares and curves) and experimental results (circles). Blue: OSIRIS, red: SILOE, circle: average of the increases of yield stress measured experimentally or determined from the DBTT shifts measured at 56 J and at 0.9 mm of lateral expansion. (a) Material M1. (b) Material M2. (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)

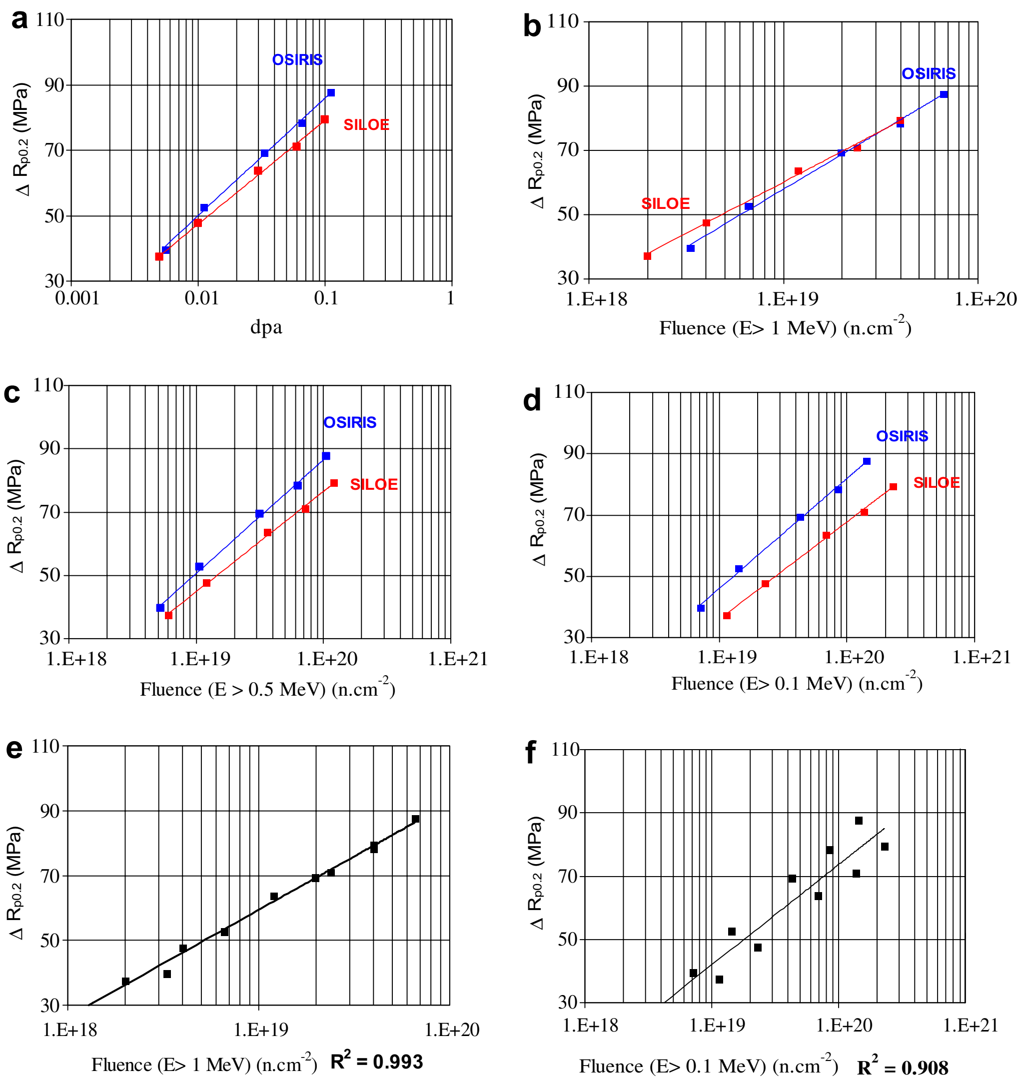

Fig. 10. Simulated increase of yield stress of material M1 after irradiation in OSIRIS or SILOE, versus several exposure parameters. (a) Same number of dpa. (b) Same fluence $E > 1$ MeV. (c) Same fluence $E > 0.5$ MeV. (d) Same fluence $E > 0.1$ MeV. (e) Trend curve relying on all simulation results obtained form the OSIRIS and SILOE spectra and plotted versus $\phi_{E>1\mathrm{MeV}}$. (f) Trend curve relying on all simulation results obtained form the OSIRIS and SILOE spectra and plotted versus $\phi_{E>0.1\mathrm{MeV}}$.

duces the higher irradiation-sensitivity of M2 compared to M1 for both neutron spectra, as observed experimentally.

- The quantitative character of the simulations is acceptable. The simulated results fall at most at 20 MPa of the experimental ones (Fig. 9).
- For both materials, the simulation with the OSIRIS spectrum induces a slightly larger increase of yield stress than with the SILOE one at the same dpa, as observed experimentally (Fig. 9).
- The simulation confirms that the spectrum effect is weak.

3.2.3. Determination of the best exposure parameter

The increases of yield stress of M1 and M2 simulated from both spectra are plotted versus $\phi_{E>1\mathrm{MeV}}$, $\phi_{E>0.5\mathrm{MeV}}$, $\phi_{E>0.1\mathrm{MeV}}$ or ϕ_{dpa} in Figs. 10 and 11. For a given material, the most appropriate exposure parameter is the one for which the trend curves drawn for both spectra are the closest. It can be observed that this condition is reached when $\Delta R_{\mathrm{P0.2}}$ is plotted versus $\phi_{E>1\mathrm{MeV}}$. The visual impression can be confirmed by calculating the regression coefficients (R^2) of the trend curve relying on all the simulated results obtained

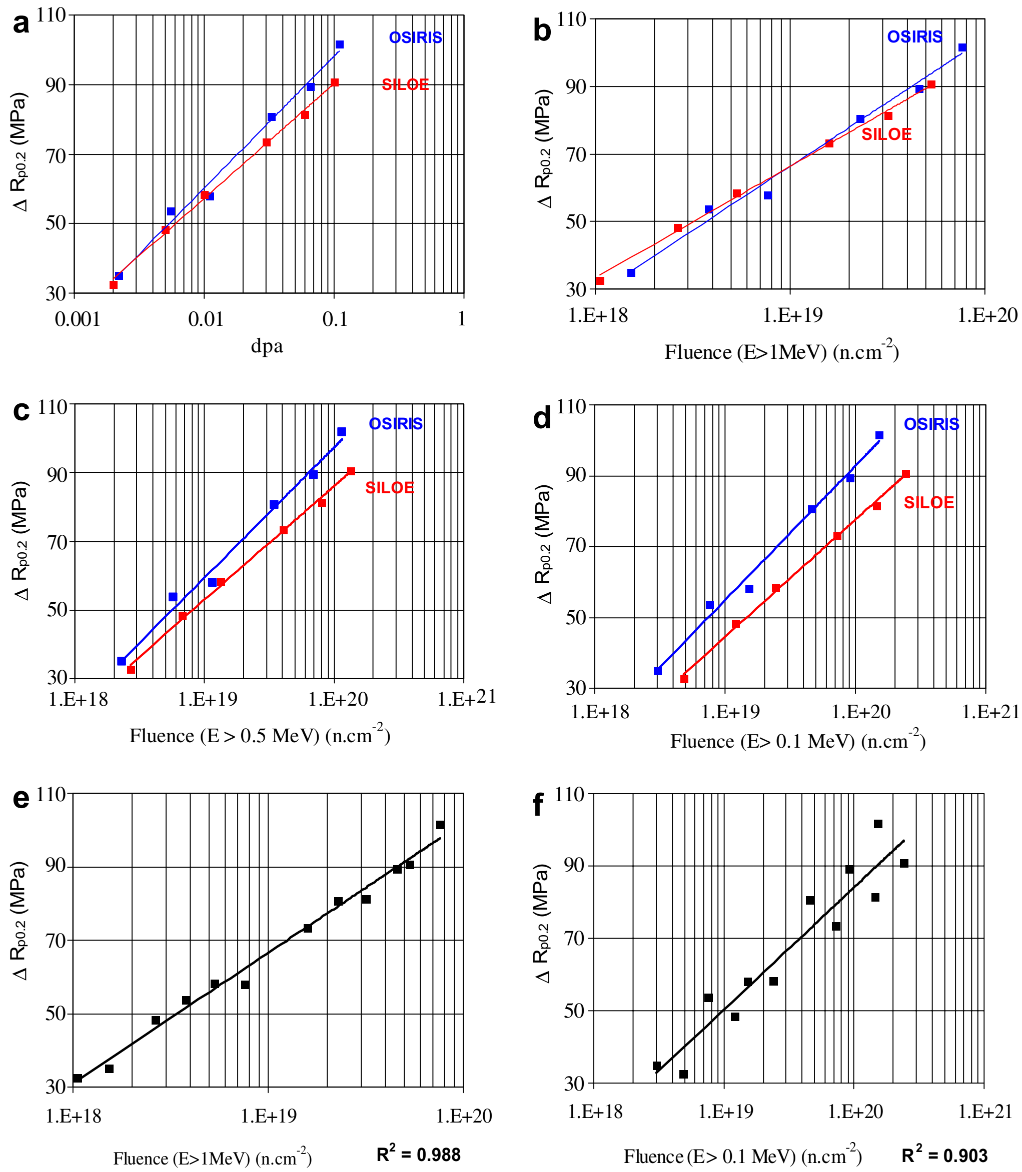

Fig. 11. Simulated increase of yield stress of material M2 after irradiation in OSIRIS or SILOE, versus several exposure parameters. (a) Same number of dpa. (b) Same fluence $E > 1$ MeV. (c) Same fluence $E > 0.5$ MeV. (d) Same fluence $E > 0.1$ MeV. (e) Trend curve relying on all simulation results obtained form the OSIRIS and SILOE spectra and plotted versus $\phi_{E>1\mathrm{MeV}}$. (f) Trend curve relying on all simulation results obtained form the OSIRIS and SILOE spectra and plotted versus $\phi_{E>0.1\mathrm{MeV}}$.

from both spectra (see as an example Figs. 10(e) and (f) as well as Figs. 11(e) and (f)). Table 8 shows that R^2 is the largest (thus the results obtained from the two spectra are the most consistent) when $\Delta R_{P0.2}$ is plotted versus $\phi_{E>1\mathrm{MeV}}$. The result obtained with the dpa is very good as well.

Despite the small differences between the two materials and between the two spectra, the simulations carried out with RPV-1 confirmed the results of the ESTEREL program. In particular, they showed that the fluence $E > 1$ MeV is a more appro-

Table 8
Effect of the exposure parameter on the regression coefficient (R^2) of the trend curves relying on all the simulation results obtained from the OSIRIS and SILOE neutron spectra

	$\phi_{E>1\mathrm{MeV}}$	$\phi_{E>0.5\mathrm{MeV}}$	$\phi_{E>0.1\mathrm{MeV}}$	ϕ_{dpa}
M1	0.993	0.964	0.9084	0.986
M2	0.988	0.955	0.903	0.979

priate exposure parameter than $\phi_{E>0.1\mathrm{MeV}}$ and ϕ_{dpa} to compare the irradiations between the surveillance capsule and the pressure vessel of the French nuclear reactors.

4. Conclusion

RPV-1 is a tool aimed at simulating the irradiation-induced increase of yield stress of reactor pressure vessel steels. It relies on many hypotheses and simplifications as well as on a poor parameterisation of the codes used to build it. Nevertheless, in the ranges of flux, fluence and material used to validate it, RPV-1 provides results which fall at about ± 20 MPa of the experimental ones. At this stage of development VTRs is not possible to get better results, which are rather satisfactory. Indeed, ± 20 MPa can be considered as the statistical dispersion of experimental irradiation-induced increases of yield stress on steels.

Furthermore, RPV-1 was used to reproduce the French experimental program ESTEREL aimed at studying the neutron spectrum effect between the surveillance capsules and the vessels of the French nuclear reactors. This simulation work led exactly to the same conclusions than those of ESTEREL, which demonstrates that RPV-1 can already been used to reinforce the conclusions of experimental programs.

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