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Fluence rate effect semi-mechanistic modelling on WWER-type RPV welds

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Abstract

Effort at JRC-IE is ongoing in order to develop a semi-mechanistic model to forecast radiation embrittlement. The understanding and the quantification of the influence of the fluence rate is of particular importance for the correct interpretation of data obtained in material testing reactors or in surveillance capsules, which are accelerated with respect to embrittlement of the reactor pressure vessel wall itself. To verify the applicability of the fluence rate as included in the semi-mechanistic model and tuning the model parameters various WWER-type vessel weld material have been studied. For the selected welds, copper ranges from 0.08 to 0.18 mass%, while phosphorus variation is from 0.013 to 0.036 mass%. The fluence range is up to 2×10^{20} n cm^{-2} obtained at two fluence rates of 4×10^{11} and 3.5×10^{12} n cm^{-2} s^{-1}, typical for WWER-440 surveillance positions. Significant fluence rate effect has been observed for the welds containing low copper and moderate phosphorus, and adaptation of the semi-mechanistic model's parameters for the high flux data is required. To verify the consistency and the limits of the findings other similar data coming from RPV surveillance programmes are also included in this analysis.

1. Background

Within the SAFELIFE action of the European Commission's Joint Research Centre-Institute for Energy (JRC-IE) the understanding of irradiation embrittlement and its mechanisms is a key subject. In the last years a semi-mechanistic model to forecast radiation embrittlement of steels has been developed [1]. This model is based on the embrittlement kinetics and takes into account not only the chemical composition and the accumulated fluence but also the irradiation temperature and the fluence rate effects. Regarding this last item, the understanding and the quantification of the effect of the fluence rate (also known as flux) is of particular importance for the correct interpretation of data obtained in material testing reactor or anyhow accelerated with respect to surveillance data or the reactor pressure vessel (RPV) wall itself.

Recently WWER-type RPV materials have been irradiated at different fluence rates by the Russian

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Research Centre, Kurchatov Institute [2] and the availability of such a high quality data offers the possibility of verifying the influence of the fluence rate as included in the semi-mechanistic model and consequently tuning the model parameters.

2. Materials, fluence and fluence rates

In order to systematically study the effect of fluence rate and its modellisation, four weld materials have been used. The materials are basically standard WWER-440 welds with different contents of phosphorous (0.013–0.036%) and copper (0.08–0.18%), their chemical composition is given in Table 1. The manufacturing process of the welds completely corresponded to standard procedure for WWER-440. These welds, which are very interesting for this type of studies, have been chosen because of the fact that for W28 and WA2 the amount of impurities and other elements does not change significantly so they can be directly used to determine the flux effect, while using W37 the additional effect of phosphorus can be investigated. Given that nickel levels are very low and the contents of manganese are practically the same for welds W28, WA2 and W37 the manganese influence is therefore analysed using weld W12.

Welds W28, WA2, W12 and W37 have been irradiated at 270 °C and two fluence rates: low fluence rate ($\mathrm{LF} = 4 \times 10^{11}\ \mathrm{n\,cm^{-2}\,s^{-1}}$ $E > 0.5$ MeV) and high fluence rate ($\mathrm{HF} = 3.5 \times 10^{12}\ \mathrm{n\,cm^{-2}\,s^{-1}}$ $E > 0.5$ MeV) and different accumulated fluences have been achieved, up to $2 \times 10^{20}\ \mathrm{n\,cm^{-2}}$. It should be noted that a fast neutron flux in the order of $3 \times 10^{11}\ \mathrm{n\,cm^{-2}\,s^{-1}}$ is a typical value of flux on the wall of WWER-440 reactor pressure vessel with full core loading.

Irradiation to high flux was carried out at Armenian NPP, Unit 2, and to low flux at Rovno NPP, Unit 1. In both cases the surveillance channel was used for the irradiation; and temperature has been kept to 270 °C, which is the typical operation temperature for WWER reactors. Typical uncertainties for surveillance channel are considered to be ±6 °C for temperature and within 12% for fluence rate determination [3].

Table 2
Range of copper and phosphorus content (mass%) for studied WWER-440 data

Material	Cu	P
SS Low flux[a]	Min: 0.03, Max: 0.12	Min: 0.01, Max: 0.023
SS High Flux[a]	Min: 0.06, Max: 0.12	Min: 0.01, Max: 0.028

[a] SS surveillance results made available by the authors.

The level of irradiation embrittlement was evaluated by the value of the ductile-to-brittle transition temperature shift (DBTT shift) obtained by standard Charpy impact tests. Actually nine Charpy V-notched specimens have been used in order to properly obtain the so-called 'impact curve'. According to the Russian Guide the transition temperature is estimated from the impact curve at a reference energy level of 47 J absorbed energy [4]. The observed DBTT shifts in the selected weld materials were as high as 220 °C.

Additional similar materials are also considered for comparison in this paper to verify consistency of the data sets and of the model fitting parameters and their limits. These materials come from relevant sub-sets (SS) of real data from WWER 440 surveillance. The cooper and phosphorus range content of such WWER-440 SS is given in Table 2.

3. Semi-mechanistic model

The data have been analysed using the semi-mechanistic model developed by the JRC-IE. Such a model is based on the general knowledge that the total irradiation embrittlement is the result of different mechanisms; including precipitation and matrix hardening [5–8]. Thus taking the additive contributions to radiation embrittlement of direct

Table 1
Relevant chemical elements for selected welds in mass%

Materials	Cu	P	Ni	Mn	Si	Cr	Mo	V	C	S
W28	0.14	0.028	0.10	1.20	0.50	1.30	0.40	0.20	0.10	0.017
WA2	0.18	0.028	0.16	1.30	0.56	1.63	0.50	0.22	0.07	0.022
W12	0.08	0.013	0.14	0.77	0.29	1.51	0.53	0.12	0.06	0.010
W37	0.13	0.036	0.15	1.32	0.20	1.11	0.38	0.20	0.06	0.011

matrix damage, precipitations and segregations [9] the total effect in term of ductile-to-brittle transition temperature shift is given in Eq. (1):

$$\mathrm{DBTT_{shift}} = a \cdot \Phi^{0.5} + b_1 \cdot \mathrm{Cu} \cdot \left[1 - \mathrm{e}^{-\Phi/\Phi_{\mathrm{sat}}}\right] + \frac{c_1 \cdot \mathrm{P}}{2} \cdot \left[1 + \tanh\left(\frac{\Phi - \Phi_{\mathrm{start}}}{d}\right) - c_0\right], \qquad (1)$$

where $\mathrm{DBTT_{shift}}$ is the transition temperature shift, Φ is the neutron fluence (10^{18} n cm^{-2}), Cu and P are the concentrations in mass% of Cu and P, respectively, a is the matrix damage parameter, b_1 is a model fitting parameter representing the maximum saturation value of the shift due to copper rich precipitations, Φ_{sat} is a model fitting parameter describing the start of saturation in the precipitation effect (the fluence at which 66% of b_1 is reached), c_1 is a model fitting parameter representing the saturation value of the shift due to segregation of phosphorus at various interfaces, Φ_{start} is a model parameter representing the fluence at which segregation starts, d is a model parameter representing the velocity of rising of $\mathrm{DBTT_{shift}}$ due to segregation until the saturation value is reached and c_0 is the balance term to force the function to be exactly zero at zero fluence, thus:

$$c_0 = \left[1 + \tanh\left(-\frac{\Phi_{\mathrm{start}}}{d}\right)\right].$$

The term c_0 is, for the obtained ratio Φ_{start}/d, normally very small, responsible of a few degrees offset, and it can be actually neglected.

This model has been developed for model alloys [10,11] irradiated in the HFR research reactor in Petten (The Netherlands), and it has been recently successfully tested on WWER-440 materials re-embrittlement [12].

Recent study to determine the effect of irradiation temperature and its inclusion in the semi-mechanistic model is published in [13].

4. Analysis of moderate Cu and high Mn welds: W28 and WA2

At first instance the semi-mechanistic model presented in Eq. (1) has been used to calculate the DBTT shifts for welds W28 and WA2. Typical coefficients a, b_1 and c_1 already obtained from analysis of similar materials and irradiation temperatures of ≈270 °C have been used to test their adequacy for the data used in this study. As Fig. 1 shows the central values are met, however it is observed that for the high flux data the calculated values of the $\mathrm{DBTT_{shift}}$ are systematically overestimated while for low flux the calculated $\mathrm{DBTT_{shift}}$ is lower than the measured.

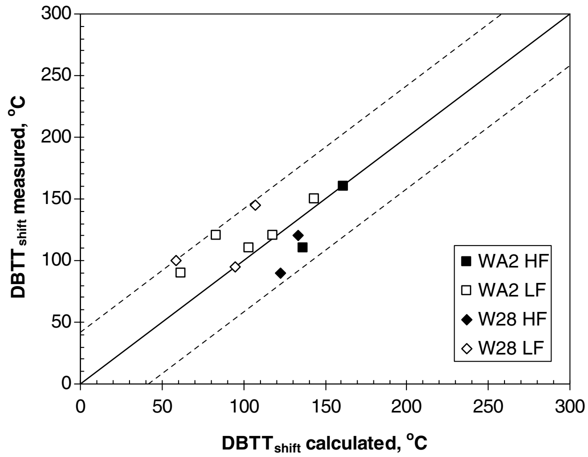

Fig. 1. Calculated versus measured $\mathrm{DBTT_{shift}}$: $a = 3$, $b_1 = 450$, $c_1 = 3000$; $\Phi_{\mathrm{start}}/d = 1$.

The largest scatter of data is found, in general, at the intermediate DBTT shifts, see Fig. 2. Such effect is clearly expected due to the basic difference between data obtained at high flux. Matrix damage is expected to increase with higher neutron fluence rates, however in the case of RPV materials (steels with a considerable amount of impurities) authors as i.e. Williams et al. in [14] have elucidated that since matrix damage is not the leading mechanism contributing to irradiation embrittlement its increase with high flux is very small in terms of DBTT shift. It

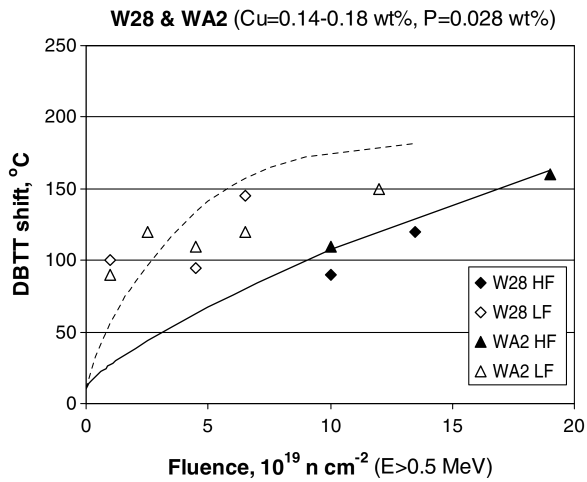

Fig. 2. Analysis of W28 and A2 welds using Eq. (1) model.

is also recognized that fluences obtained at very high fluence rates are accumulated in such a short time that might be shorter than the typical times required for fully developing other processes as precipitation or segregation [14].

In fact diffusion and precipitation's formation are controlled in a given irradiation temperature by the irradiation time. This effect appears in the fluence-$DBTT_{shift}$ plot as different ways to reach saturation. A clear example can be observed in Fig. 2 for the W28 and WA2 data sets. In Fig. 2 the two welds are analysed together as the content of phosphorus is the same (P = 0.028 mass%) and the copper content does not vary significantly (Cu = 0.14–0.18 mass%). For the same accumulated fluence if the fluence rate is higher the obtained DBTT shifts are smaller. According to the semi-mechanistic model the consequent assumption is that the fluence rate influences the model constants describing saturation and initiation of precipitation and segregations: the parameters Φ_{sat} and Φ_{start} of Eq. (1) that can be considered as 'time-dependent'.

Manganese is also affecting the precipitation process; in particular for high nickel steels [15–17], but in the case of W28 and WA2 this effect cannot be distinguished since the amount of Mn is almost the same for both welds and the Ni content is relatively low.

The curves drawn in Fig. 2 are given by Eq. (1). The dotted curve has been obtained using standard values for Φ_{sat} and Φ_{start}, valid generally for low flux, while the solid line is obtained adapting the value of the parameters for the higher flux data.

Significant adjustments of both Φ_{sat} and Φ_{start} as a function of the fluence rate are required in order to reproduce the observed data, see Fig. 3.

As can be seen in Fig. 4, the inclusion of the flux correction factor(s) allows better distribution of $DBTT_{shift}$ values along the median and reduces the data scattering.

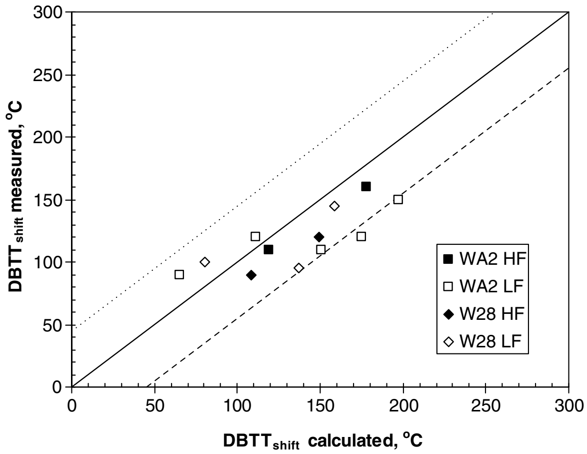

Fig. 4. Calculated versus measured $DBTT_{shifts}$ adjusted Φ_{sat} and Φ_{start} for high flux.

5. Analysis of low Cu and low Mn weld: W12

Weld W12 contains the lowest copper amount (very near to estimated Cu threshold precipitation effects [18,19]) among all analysed welds (Cu = 0.08 mass%) and moderate content of phosphorus (P = 0.013 mass%). In addition the level of manganese is also lower than the previous welds (Mn = 0.77 mass%). It can be noted in Fig. 5 that no significant fluence rate effect on $DBTT_{shift}$ is observed for W12.

6. Analysis of high Mn and very high P weld: W37

From what we have observed so far on welds with a high content of manganese (Mn ≈ 1.32 mass%) a

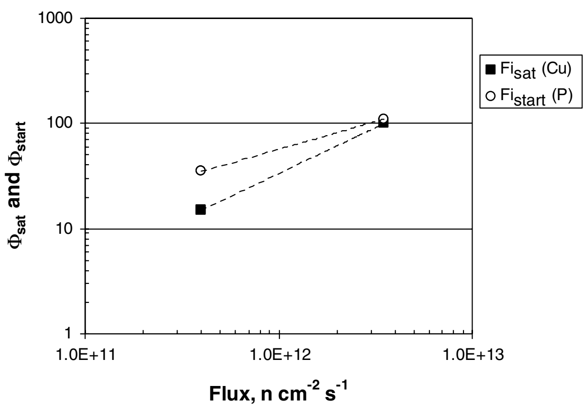

Fig. 3. Trend of required for Φ_{sat} and Φ_{start} parameters (in 10^{18}).

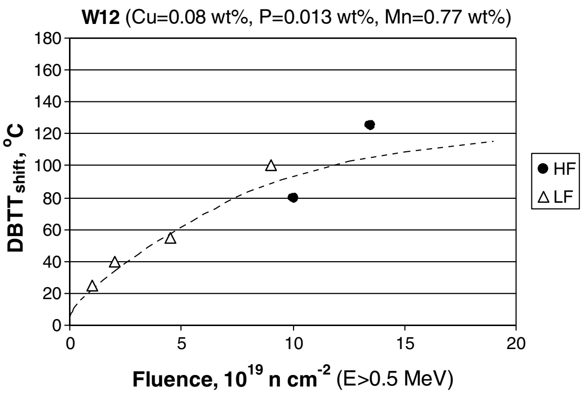

Fig. 5. Analysis of W12 weld using Eq. (1) model.

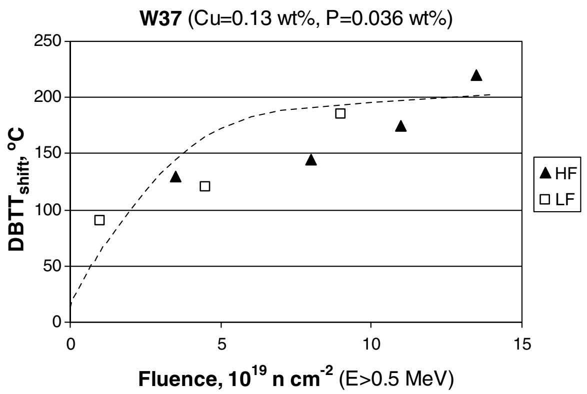

Fig. 6. Analysis of W37 weld using Eq. (1) model.

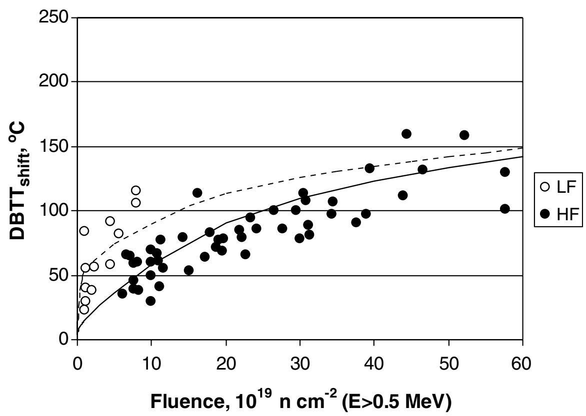

Fig. 7. Analysis of WWER-440 surveillance data (welds) using Eq. (1) model.

fluence rate effect exists. However in Fig. 6 can be noted that the situation is not the same for weld W37. Weld 37 contains also moderate copper like the welds W28 and WA2 but the phosphorus content in much higher (P = 0.036 mass%).

Little adaptation of Φ_{sat} and Φ_{start} parameters should be required in order to reproduce the observed data at high flux which are not showing any different trend behaviour than those obtained at low flux. The trend-curve drawn in Fig. 6 is in fact obtained by using Eq. (1) and values Φ_{sat} and Φ_{start} typical of low fluence rate.

7. Discussion

The analysis of the studied welds is showing that significant adaptation of the 'time-dependent' parameters, Φ_{sat} and Φ_{start}, is required in order to properly model the DBTT shifts obtained at low flux and at high flux. This is particularly true for data from low to intermediate fluence levels. At higher accumulated fluences the flux effect is less significant since saturation effects occur.

In general, for the moderate copper regimes typical of the analysed data, a clear need to use higher values of Φ_{sat} fluence (which represents the beginning of saturation in the copper precipitation effect) for high flux data has been identified. Furthermore, when copper precipitation is leading the embrittlement, manganese seems to shorten the time to complete that precipitation process, which is consequently modelled lowering the value of Φ_{sat}.

In the case of the higher phosphorus weld, segregation of P leads the embrittlement and the observed effect of flux is negligible.

To verify the consistency of the semi-mechanistic model, relevant surveillance data from WWER-440 reactors, see Table 2, irradiated in different positions (hence at different fluence rates) are also added to the present analysis. The data show some scattering due to the variations within the limits of Cu and P, and also due to the fact that the data cannot be discriminated per Mn level. Although the manganese contents for individual data are not available, in Ref. [20] it can be seen that the data are in the moderate Mn range with content less than 0.8 mass%.

In spite of the scattering, a global agreement with the already discussed results has been found. Irradiations at high flux are in fact producing smaller DBTT shifts in the low fluence range as Fig. 7 shows.

The possible explanation of the apparent increase of Φ_{sat} when modelling embrittlement at high flux, can be found if we consider that many physical phenomena are in fact time dependent in a given temperature; in particular diffusion and clustering processes towards formation of precipitations. The completion of such processes is a priori dependent on neutron fluence, while the production rate of defects would affect their kinetics. In this way fluence accumulated at higher fluence rate might be achieved in such a short time that might not be enough to finalize the above-mentioned processes. The situation is shown, as example, in Fig. 8 in which the irradiation time is drawn versus the accumulated fluence. If we assume a minimum time to complete precipitation processes, t_{min}, there would be a region for fluences (the part below t_{min}) where the damage component related to precipitation cannot be concluded and hence its effect on DBTT shift is only partial. This means a lower visible DBTT$_{shift}$.

If the time to achieve a certain fluence level, even at high flux, is higher than the minimum time the

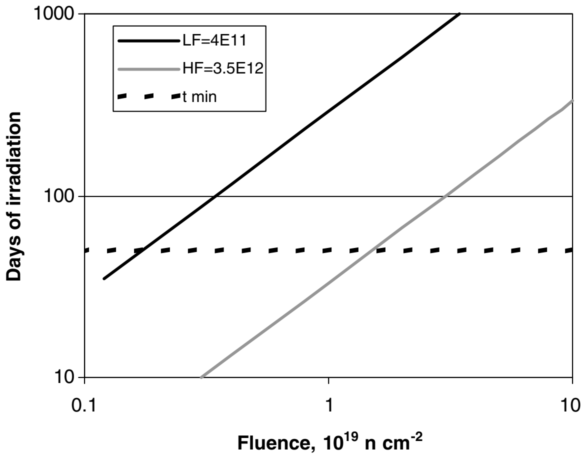

Fig. 8. Times to accumulate fluence at different fluence rates.

copper precipitation contribution to DBTT shift would be complete.

The proposed semi-mechanistic model can be then used to determine the necessary correction term, FF, to adjust the shifts obtained at high flux, $\mathrm{DBTT}^{\mathrm{HF}}_{\mathrm{shift}}$. The factor FF is calculated as the required value to add to $\mathrm{DBTT}^{\mathrm{HF}}_{\mathrm{shift}}$ in order to find the real shifts at low flux, $\mathrm{DBTT}^{\mathrm{LF}}_{\mathrm{shift}}$.

$$\mathrm{FF} = \mathrm{DBTT}^{\mathrm{LF}}_{\mathrm{shift}} - \mathrm{DBTT}^{\mathrm{HF}}_{\mathrm{shift}}. \tag{2}$$

Using Eq. (1) and re-arranging we can then write

$$\mathrm{DBTT}^{\mathrm{LF}}_{\mathrm{shift}} = a \cdot \Phi^{0.5} + b_1 \cdot \mathrm{Cu} \cdot \left[1 - \mathrm{e}^{-\Phi/\Phi^{\mathrm{LF}}_{\mathrm{sat}}}\right] + \frac{c_1 \cdot \mathrm{P}}{2} \cdot \left[1 + \tanh\left(\frac{\Phi - \Phi^{\mathrm{LF}}_{\mathrm{start}}}{d}\right)\right], \tag{3}$$

$$\mathrm{DBTT}^{\mathrm{HF}}_{\mathrm{shift}} = a \cdot \Phi^{0.5} + b_1 \cdot \mathrm{Cu} \cdot \left[1 - \mathrm{e}^{-\Phi/\Phi^{\mathrm{HF}}_{\mathrm{sat}}}\right] + \frac{c_1 \cdot \mathrm{P}}{2} \cdot \left[1 + \tanh\left(\frac{\Phi - \Phi^{\mathrm{HF}}_{\mathrm{start}}}{d}\right)\right], \tag{4}$$

where $\Phi^{\mathrm{LF}}_{\mathrm{sat}}$ and $\Phi^{\mathrm{HF}}_{\mathrm{sat}}$ are the saturation parameters at low and high flux, respectively; and $\Phi^{\mathrm{LF}}_{\mathrm{start}}$ and $\Phi^{\mathrm{HF}}_{\mathrm{start}}$ are the segregation parameters at low and high flux, respectively.

Substituting in Eq. (2) we obtain:

$$\mathrm{FF} = b_1 \cdot \mathrm{Cu} \cdot \left[-\mathrm{e}^{-\Phi/\Phi^{\mathrm{LF}}_{\mathrm{sat}}} + \mathrm{e}^{-\Phi/\Phi^{\mathrm{HF}}_{\mathrm{sat}}}\right] + \frac{c_1 \cdot \mathrm{P}}{2} \cdot \left[\tanh\left(\frac{\Phi - \Phi^{\mathrm{LF}}_{\mathrm{start}}}{d}\right) - \tanh\left(\frac{\Phi - \Phi^{\mathrm{HF}}_{\mathrm{start}}}{d}\right)\right]. \tag{5}$$

For low phosphorus materials, or for all cases where mainly copper precipitation is influenced by flux, or when $\Phi^{\mathrm{LF}}_{\mathrm{start}} \approx \Phi^{\mathrm{HF}}_{\mathrm{start}}$ Eq. (5) can be simplified as

$$\mathrm{FF} = b_1 \cdot \mathrm{Cu} \cdot \left[\mathrm{e}^{-\Phi/\Phi^{\mathrm{HF}}_{\mathrm{sat}}} - \mathrm{e}^{-\Phi/\Phi^{\mathrm{LF}}_{\mathrm{sat}}}\right]. \tag{6}$$

The FF factors calculated using Eq. (6) as a function of fluence are shown in Fig. 9 for different copper contents. The parameters $\Phi^{\mathrm{LF}}_{\mathrm{sat}}$ and $\Phi^{\mathrm{HF}}_{\mathrm{sat}}$ applied in Eq. (6) are those typical for the materials considered in this study.

Therefore FF can be used to take into account the fluence rate effect, making possible to estimate the actual value of the ductile-to-brittle transition temperature shift of a RPV in a given accumulated fluence by means of accelerated data (i.e. data obtained at higher flux coming from irradiations in surveillance capsules or at material testing reactors). As can be seen in Fig. 9 at low fluences the FF could be particularly relevant, especially for

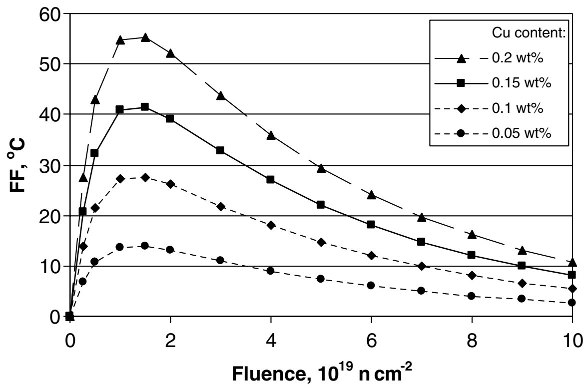

Fig. 9. FF factor versus fluence.

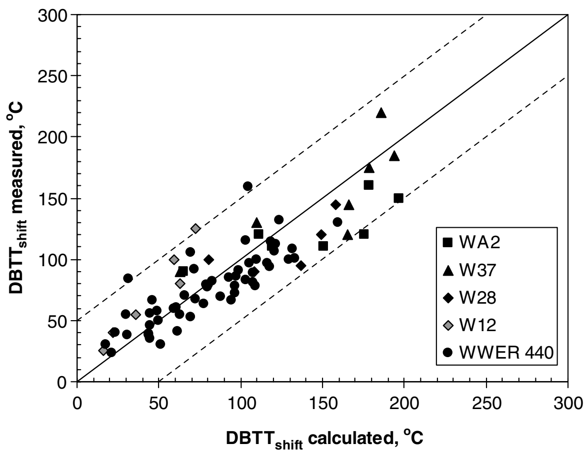

Fig. 10. Calculated versus measured $\mathrm{DBTT_{shifts}}$ for all data.

materials with high copper content, while for high fluences (approaching end-of-life) the correction factor is becoming less important.

In Fig. 10 the prediction capabilities of the corrected semi-mechanistic model are shown for the data analysed in this paper.

Finally it should be mentioned that, in the case of welds for which copper levels are low or negligible and phosphorus contents high enough to allow segregation being the dominant embrittlement mechanism, more data and research are required in order to establish a proper fluence rate dependence.

8. Conclusions

The semi-mechanistic model to forecast radiation embrittlement kinetics developed by JRC-IE is used in this article to analyse the recently produced data at RRC-KI on WWER-type RPV welds. The studied welds have been irradiated at different fluence rates and are used in this work to verify the model assumptions and to consequently tune the relevant parameters.

Significant fluence rate effects can be observed for the low to intermediate copper regime and up to moderate phosphorus contents. For the same accumulated fluence the obtained DBTT shifts are smaller when the fluence rate is higher. The largest scatter of data is found for the intermediate DBTT shifts; which is consistent with the hypothesis that fluences obtained at very high fluence rates may have be obtained in such a short time that might be shorter than the time required for other processes (diffusion, clustering and precipitates formation), to fully develop. A significant adaptation of the parameters modelling precipitation and segregation for the high flux data is therefore required in order to reproduce the observed data.

To verify the consistency of the findings WWER welds surveillance data have also been included in this study. A correction term taking into account the flux effect when estimating the value of the DBTT shift of a RPV from accelerated data appears to be relevant for low accumulated fluences.

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