Heliyon 9 (2023) e16581

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

The development of prediction model on irradiation embitterment for low Cu RPV steels

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ARTICLE INFO

Keywords:
RPV
Irradiation embrittlement
Physical mechanism
Prediction model

ABSTRACT

The development of prediction model on irradiation embitterment (PMIE) of reactor pressure vessel (RPV) is an important method for nuclear reactor long term operation. Based on the physical mechanism of RPV irradiation embrittlement, a preliminary model is determined and the critical threshold of Cu content of 0.072% is obtained according to this preliminary model. Then a prediction model named PMIE-2020 for low Cu RPV steels is developed. At last the residual, standard deviation and predicted values and test values distribution analysis are given. Simultaneously, a comparison between PMIE-2020 and other prediction model and irradiation data is provided. Results indicate that the predicted results of PMIE-2020 has no tendency with influence factors such as neutron fluence, flux, irradiation temperature, chemical elements Cu, P, Mn, Ni, Si. The residual standard deviation is 10.76 °C, which is lower than present prediction model. The distribution between predicted values of PMIE-2020 and test values are located the area near the 45° line. These results prove that the PMIE-2020 have high accuracy on irradiation embrittlement prediction.

1. Introduction

1.1. Background

Reactor pressure vessel (RPV) is the pressure boundary of the primary water and cannot be replaced during service lifetime. Its safe working time has become the key to determining the life of the nuclear power plant. RPV is irradiated by fast neutrons ($E > 1$ MeV) during the service period and irradiation embrittlement phenomenon is considered as the most important failure mechanism [1]. Irradiation embrittlement will cause low-stress rupture of in-service RPV, which directly threatens the safety of nuclear power plants.

Although the ΔRT_{NDT} (reference zero plastic transformation temperature increment) of RPV steel can be obtained by impact test of irradiation surveillance sample, it is not possible to evaluate irradiation embrittlement level continuously and obtain the embrittlement tendency [2]. So irradiation embrittlement prediction of RPV steel is fundamental in ensuring safe operation of nuclear reactors. Therefore, development of prediction model on irradiation embrittlement (PMIE) for RPV steel has realistic engineering application requirements [3]. Under this background, several PMIE have been developed in past 30 years, such as US RG1.99 (Rev. 2) [4], NUREG/CR-6551 [5] and ASTM E900-02 [6], France RCC-M ZG3430 [7] and RSEM B7213 [8], Japan JEAC 4201 [9], etc.

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https://doi.org/10.1016/j.heliyon.2023.e16581
Received 10 July 2022; Received in revised form 19 May 2023; Accepted 20 May 2023
Available online 23 May 2023

RPVs are usually made of ferritic MnNiMo steel. Early nuclear power plants used A302B steel, some of which are still in operation. Since 1960s and 1970s, forged A508-2 steel and its modified grade A508-3 steel are mainly used as RPV steel. No matter A302B, A508-2 or A508-3 steel, there are different levels of Cu due to the smelting cost and understanding of Cu on irradiation embrittlement. So large amount of high Cu content RPV embrittlement data was obtained and used. Based on these high Cu irradiation data, many prediction models were developed. However, the accuracy of these prediction model under low Cu content condition may deviate from the actual situation. So these early models cannot meet the embrittlement prediction and assessment of low Cu RPV steel.

On the other hand, previous irradiation supervision data and experimental reactor data corresponded to a relatively short operating time (smaller fluence range) of nuclear power plants [3,10]. With the further development of nuclear power technology, long term operation and life extension of nuclear power plants have become a new requirement. Therefore, neutron fluence will inevitably increase with the increase of operation time and thus exceed the applicable scope of previous prediction model. Moreover, with the development of embrittlement mechanism, new embrittlement influencing factors (for example, the influence of Ni–Mn-(Si) precipitates on embrittlement at high fluence condition) have been recognized [10,11]. New embrittlement mechanism should be added into the prediction model. Therefore, developing a new and more accurate PMIE is an actual needs.

1.2. RPV irradiation embrittlement mechanism

Irradiation embrittlement mechanism is the basis of PMIE development. Two main types of microstructural features, namely solute clusters (especially the copper-rich clusters and Mn–Ni-(Si) clusters) and matrix damage (especially the dislocation loops), have been considered to be the key embrittlement mechanism [12]. The main parameters of PMIE should be the influencing factors of irradiation embrittlement. Previous PMIEs, such as RG1.99 (Rev. 2), RCC-M ZG3430, RSEM B7213 and JECA 4201, can be expressed as $\Delta RT_{NDT} = CF \times FF$, where CF is the influence factor of chemical composition, FF is the influence factor of irradiation fluence. With the deeply understanding of irradiation embrittlement mechanism on RPV steel, new PMIEs, such as NUREG/CR-6551 and ASTM E900-02, are developed and can be expressed as $\Delta RT_{NDT} = \Delta T_{MD} + \Delta T_{CRP}$, where ΔT_{MD} is the temperature increment caused by matrix defects (MD), and ΔT_{CRP} is the temperature increment caused by Cu-related phase (CRP).

MD mainly originates from neutron cascade collisions during the irradiation process, including dislocation loops, vacancy-solute cluster complexes or their solute (Ni, Mn and Si) remnants, etc [13]. ΔT_{MD} is negatively related to the irradiation temperature and approximately proportional to the square root of the neutron fluence. The negative correlation between ΔT_{MD} and temperature is due to the thermal stability of MD. So MD is more stable under low temperature conditions. Meanwhile, The square root relationship between ΔT_{MD} and neutron fluence is directly related to the irradiation hardening, which mainly caused by dispersed barriers to dislocation glide [14]. CRP is mainly composed of Cu-rich phase and Mn–Ni-(Si)-rich phase, which is directly related to the vacancy concentration during irradiation. Cu-rich phase only appears when the Cu element content in RPV steel exceeds a certain critical threshold [10]. If the Cu content in RPV steel is larger than this critical threshold, it is called high Cu RPV steel, otherwise, it is called low Cu RPV steel. Mn–Ni-(Si) phase can be formed in high Cu or low Cu RPV steel. In high Cu RPV steel, the formation of Mn–Ni-(Si) phase depends on the Cu-rich phase. In low Cu RPV steel, Mn–Ni-(Si) phase has nothing to do with Cu (proved by thermodynamic and kinetic models) [15,16] and is more pronounced under high irradiation damage conditions. So Mn–Ni-(Si) phase in low Cu RPV steels is called late blooming phases (LBP). Although the formation of LBP has nothing to do with Cu content, its formation mechanism is still irradiation-enhanced diffusion of solute atoms, which is similar to the Cu-rich phase. So in this study LBP is still classified as CRP.

2. Irradiation embrittlement data

The PMIE is developed according to irradiation embrittlement data. So the number of irradiation embrittlement data determines the reliability of the PMIE to a certain extent. In this studies, the irradiation supervision data and experimental reactor data are

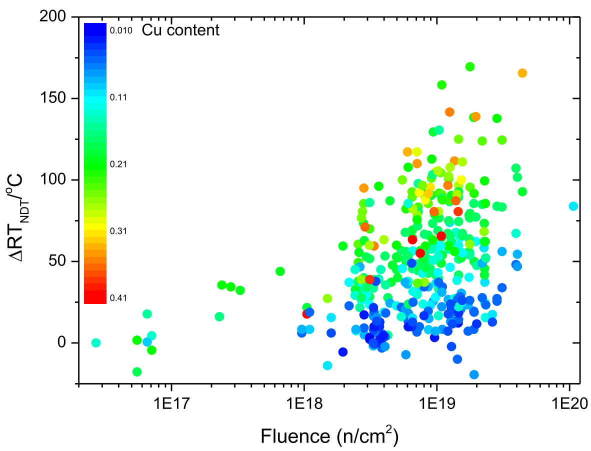

Fig. 1. The variation of ΔRT_{NDT} vs. neutron fluence and Cu content.

collected from French, America, Japan and China [17]. The data subjects include material type, chemical composition (including Cu, P, Mn, Ni, Si, etc.), neutron fluence, flux, irradiation temperature, ΔRT_{NDT}, etc. The correlation between neutron fluence, Cu content and ΔR_{TNDT} of some data is shown in Fig. 1.

2.1. 3Model development

Fig. 2 is the flow chart of PMIE development. Based on the irradiation embrittlement data and preliminary model selected, the Cu threshold can be determined and thus the low Cu data used in this study can be screened out. It should be noted that the preliminary model should contain the neutron fluence parameter because the fluence is the most important and significant influence factor according to Fig. 1. After parameters modification of this preliminary model, other influencing factors can be added according to the irradiation embrittlement physical mechanism and corresponding parameters can be obtained. After the PMIE establishment, the statistical analysis, comparative analysis and irradiation and material variables analysis is given to verify the accuracy and reliability of this model.

Previously a relationship between ΔRT_{NDT} and neutron fluence and irradiation temperature is provided by Jones et al. [18]. The expression is as follow:

$$\Delta RT_{NDT} = A_0(1 - 0.002445T) \cdot f^{0.5} \quad (1)$$

where the T is the irradiation temperature with ^{o}C, f is neutron fluence with n/cm^2, A_0 is the coefficient. On the other hand, according to the aspect of hardening embrittlement, the hardness is also approximately proportional to the square root of the neutron fluence (such as Orowan model) [19]. Moreover, in previous PMIEs, ΔRT_{NDT} is approximately proportional to the square root of the neutron fluence, namely $\Delta RT_{NDT} \infty f^{0.5}$. These PMIE include RCC-M ZG3430 model, NUREG/CR-6551 model and ASTM E900-02 model, whose ΔRT_{NDT} are proportional to the index of 0.5, 0.4601 and 0.5076 [5–7]. Therefore, an approximate square root relationship between ΔRT_{NDT} and neutron fluence is a universally accepted conclusion. Therefore, according to present understanding on irradiation embrittlement mechanism and earlier PMIE, equation (1) can be used as the preliminary model of the PMIE developed in this study.

It can be observed from Fig. 1 that the irradiation embrittlement (ΔRT_{NDT}) of RPV steel increases significantly with the increase of Cu content. Further analysis of the irradiation embrittlement mechanism shows that there is a threshold for chemical element Cu on irradiation embrittlement effect. If the Cu content is lower than this threshold, the chemical element Cu has no effect on irradiation embrittlement [20]. In order to determine the threshold of Cu, the data with Cu content less than 0.13% in the irradiation embrittlement data are grouped, and the parameter A_0 and the average residual (the residual is the difference between predicted and tested value; the average residual is the mean residual in each Cu group) varies with the Cu content are calculated according to equation (1) as shown in Fig. 3. The results show that when Cu $\leq$ 0.07%, the parameter A_0 and the average residual obtained according to the preliminary model (equation (1)) do not change significantly with the increase of Cu content, and $A_0 = 1.954 \times 10^{-8}$. When the Cu content is greater than 0.07%, the parameter A_0 and the average residual are significantly correlated with the increase of Cu content. So the threshold for chemical element Cu on irradiation embrittlement effect according to irradiation data is 0.07%. Moreover, according to the latest irradiation embrittlement understanding on Cu [6,14], this Cu threshold is revised to 0.072%. Therefore, when Cu $\leq$ 0.072%, the irradiation embrittlement of RPV steel has nothing to do with Cu. The preliminary model (equation (1)) on irradiation embrittlement of low Cu RPV is revised as

$$\Delta RT_{NDT} = 1.954 \times 10^{-8}(1 - 0.002545T) \cdot f^{0.5} \quad (2)$$

According to the Cu threshold (0.072%), the low Cu RPV irradiation embrittlement data can be screened. Then residual analysis is

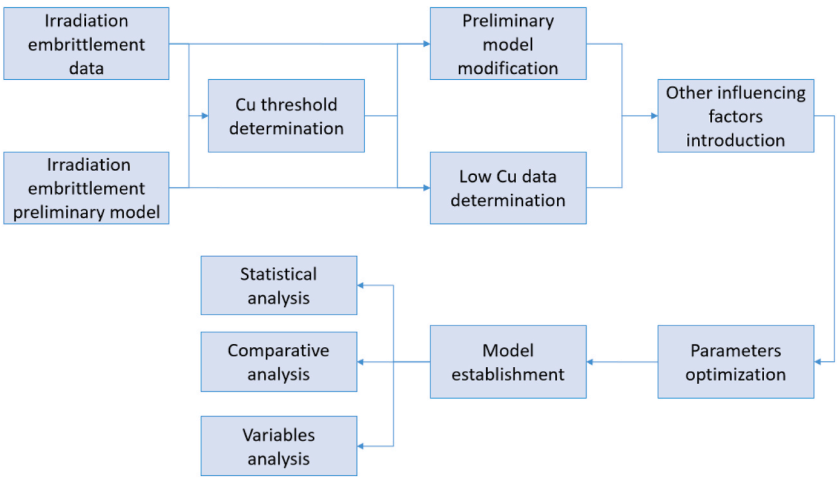

Fig. 2. The flow chart of PMIE development.

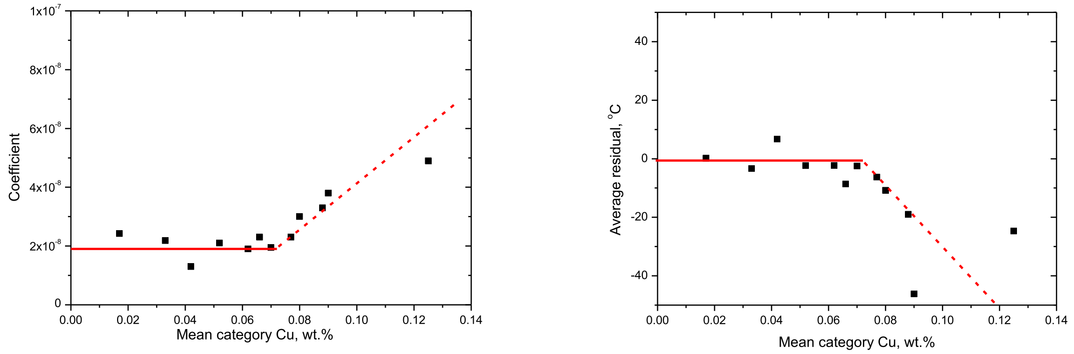

Fig. 3. The variations of A_0 (left) and average residuals (right) with the Cu content according to equation 1.

performed based on the preliminary model (equation (2)). The results (not shown here) show that the residual curve deviates greatly from the residual zero line, and the residual curve tends to change with the increase of Mn, Ni, and Si content, indicating that the preliminary model (equation (2)) does not fully consider the irradiation embrittlement factors and irradiation embrittlement data are not fully reflected. Mn, Ni and Si content show direct effect on irradiation embrittlement. So the preliminary model (equation (2)) needs to consider the influence factors of Mn, Ni and Si on irradiation embrittlement. Moreover, previous microstructural analysis indicated that LBP will be formed in RPV steel under high fluence condition and cause the irradiation embrittlement effect. The ΔRT_{NDT} and Mn–Ni–Si phase fraction show a power-law dependence according to the studies by NUGENIA Association [21] and Orowan model. Meanwhile, the PMIE developed by Eason et al. [14] considered the influence of Mn on irradiation embrittlement in the non-Cu influence related terms. Refer to the form of Mn in this model, the influence of Mn, Ni, Si on the irradiation embrittlement are introduced, and equation (2) can be modified to include Mn, Ni, Si term as shown in equation (3).

$$\Delta RT_{NDT} = 1.954 \times 10^{-8}(1 - 0.002545T)f^{0.5}(1 + B \cdot Mn^{x} Ni^{y} Si^{z}) \tag{3}$$

where the Mn, Ni, Si is the content with wt.%, B, x, y, z is corresponding parameters.

With low Cu RPV irradiation embrittlement data, using the computer optimal solution analysis, the optimal parameters B, x, y, z were obtained. Then minor adjustments on fluence index were made and the bias coefficient was introduced. So a low Cu (Cu < 0.072 wt%) RPV steel irradiation embrittlement prediction model PMIE-2020 is developed and the model form is as equation (4).

$$\Delta RT_{NDT} = 1.954 \times 10^{-8}(1 - 0.002545T)\left(1 + 0.3Mn^{2.2}Ni^{0.15}\right)f^{0.492} + 1.9 \tag{4}$$

where ΔRT_{NDT} is the reference zero plasticity transition temperature increment, with °C. The symbol and usage scope for PMIE-2020 is shown in Table 1.

3. Analysis and discussion

3.1. Statistical analysis

Fig. 4 shows the relationship between the residual of the PMIE-2020 and the neutron fluence, irradiation temperature, chemical elements Cu, P, Mn, Ni, and Si. The residual curve is calculated by the least square method. The results show that the residual curve is almost at the residual zero line, and there is no significant tendency, indicating that the PMIE-2020 has high accuracy and the ΔRT_{NDT} can be described by irradiation fluence, irradiation temperature, Mn, and Ni content.

The standard deviation of PMIE-2020 is 10.81 °C, which is smaller than the standard deviations of RG1.99 (Rev. 2) (14.8 °C), NUREG/CR-6551 (12.8 °C), ASTM E900-02 (12.2 °C), and also lower than Mark model (11.9 °C) [22] and EONY model [14] (10.88 °C), indicating that the accuracy of PMIE-2020 is slightly higher than the current prediction model. Fig. 5 shows the distribution of PMIE-2020 predicted and tested values. It can be seen that the predicted value of PMIE-2020 has good consistency with the tested

Table 1
The symbol and usage scope of PMIE-2020.

Parameter	Symbol	Usage scope	Unit
Neutron fluence (E > 1 MeV)	f	1×10^{19}–10×10^{19}	n/cm^2
Flux	Φ	5×10^{8}–2×10^{11}	$n/cm^2 \cdot s$
Temperature	T	275–292	°C
Mn content	Mn	0.58–1.83	Wt.%
Ni content	Ni	0.065–0.973	Wt.%
P content	P	0.003–0.02	Wt.%
Cu content	Cu	≤0.072	Wt.%

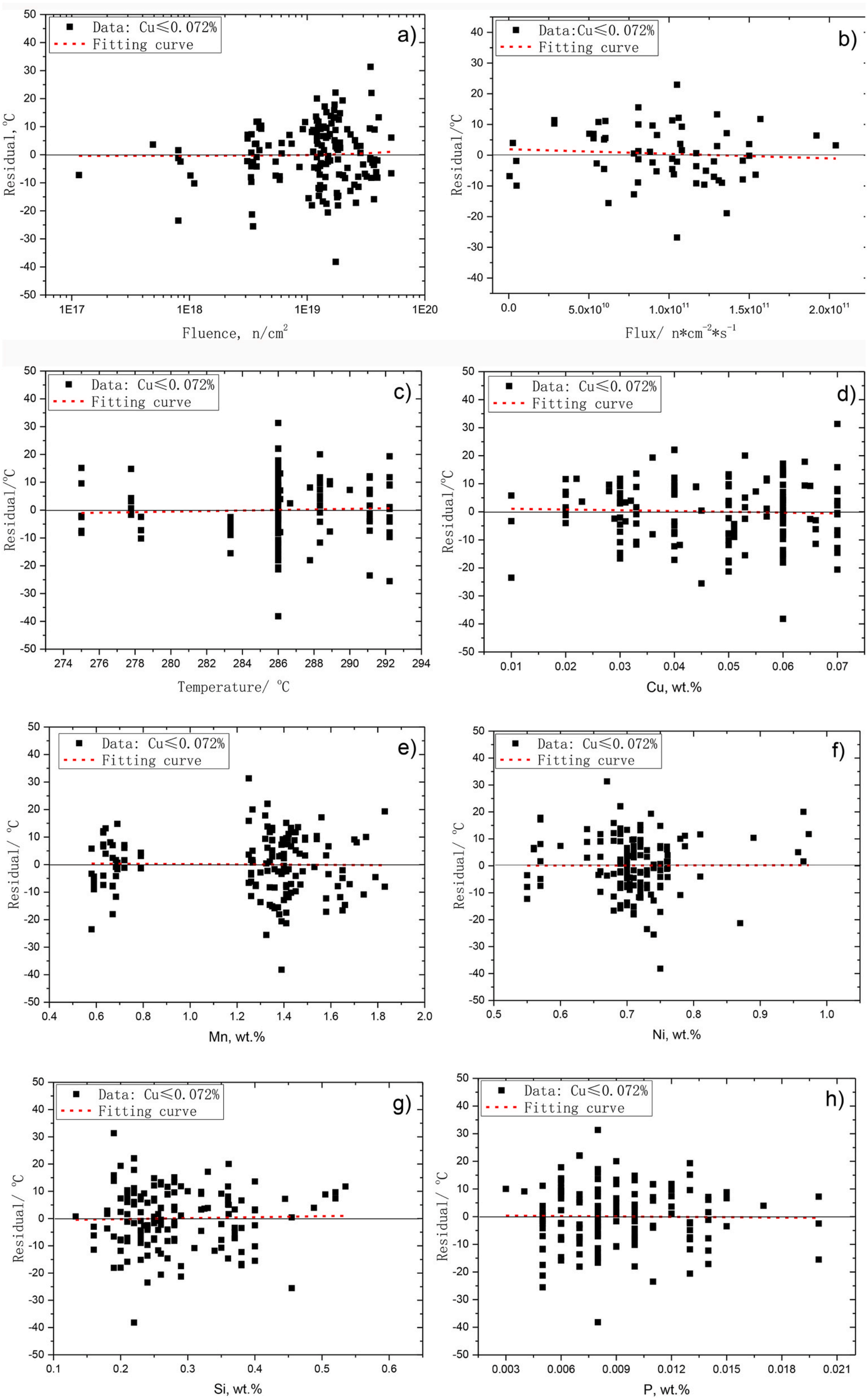

Fig. 4. The residual with a) neutron fluence, b) neutron flux, c) irradiation temperature, d) Cu, e) Mn, f) Ni, g) Si, h) P based on PMIE-2020.

value. Most of the data points fall in the area near the 45° line and basically within the 95% confidence interval of PMIE-2020, indicating PMIE-2020 has high accuracy and reliability.

3.2. Comparative analysis

Fig. 6 shows the comparison of PMIE-2020 and other prediction models with irradiation data. The results show that the FIS model (RSEM-1990) is basically near the upper envelope of the irradiation embrittlement data, showing a significant conservatism. Due to the limitation of use range, FIM-1990 cannot predict the irradiation embrittlement under high fluence conditions. Under lower fluence condition, the prediction results of PMIE-2020 are basically consistent with the RG1.99 (Rev. 2) and RSEM (2010) models. Under high fluence condition, with the increase of neutron fluence, the prediction trend of PMIE-2020 is gradually separated from the prediction results of RG1.99 (Rev. 2) and RSEM (2010) models. This is because RG1.99 (Rev.2) is based on the early neutron irradiation data and does not consider the irradiation damage mechanism under high fluence conditions. Although the usage scope of RSEM (2010) has been expanded to $11.5 \times 10^{19} n/cm^2$, The prediction results are too conservative on low Cu data. PMIE-2020 is in good agreement with the irradiation data. Especially under higher fluence condition, due to the LBP on irradiation embrittlement, PMIE can accurately reflect the irradiation embrittlement trend.

3.3. Irradiation and material variables analysis

The neutron flux is an important factor affecting the irradiation hardening and embrittlement of RPV steel. A typical 1000 MW commercial reactor has a neutron flux level of less than $6 \times 10^{10}\ n/cm^2 \cdot s$. Previous studies [23–25] indicates that, in low Cu RPV steel, under the condition of neutron flux $<1 \times 10^{12}\ n \cdot cm^{-2} \cdot s^{-1}$, neutron flux has no significant effect on hardening embrittlement. Compared with other factors, MD is the main factor causing hardening and embrittlement in low Cu RPV steel, thus the MD parameters (defect concentration and size etc.) have become the key factors affecting hardening and embrittlement. Although defect concentration gradually increases with the increase of the flux within a certain flux range, with the increase of the defect mobility, the probability of self-annihilation of vacancies and interstitial atoms increases. This will lead to the defect concentration remains basically unchanged. Therefore, in low Cu RPV steel, when flux is less than $1 \times 10^{12}\ n \cdot cm^{-2} \cdot s^{-1}$, there is no need to consider the hardening and embrittlement caused by flux. In this study, the flux range of PMIE-2020 is small than $1 \times 10^{12}\ n \cdot cm^{-2} \cdot s^{-1}$. According to the analysis above, in the process of establishing the PMIE, There is no need to consider the neutron flux effect. Fig. 7 shows the residual standard deviation of PMIE-2020 in different flux ranges. It is indicated that in the range of $2.6 \times 10^9\ n \cdot cm^{-2} \cdot s^{-1}$ to $2 \times 10^{11}\ n \cdot cm^{-2} \cdot s^{-1}$, the residual standard deviation of PMIE-2020 is between 5.6 and 10.2 °C. The residual standard deviation varies little with flux. In addition, the residual curve in Fig. 4 does not have a significant tendency with the change of neutron flux and is distributed near the residual zero line. Therefore, PMIE-2020 does not consider the effect of neutron flux.

The chemical element P will segregate at the sub-interface or grain boundary (lead to non-hardening embrittlement) or from phosphide phases (lead to hardening embrittlement). The effect of P on irradiation embrittlement has different understanding. Some studies [14] show that the effect of P on irradiation embrittlement is significantly weakened when the Cu content is reduced. However, Nikolaev et al. [26] suggest that the effect of phosphorus content on RPV steel with low copper content can be very significant. However, in present study, there is no clear tendency change of residual with the change of P content (Fig. 4), indicating that present low Cu irradiation embrittlement data cannot confirm the effect of P on irradiation embrittlement. This is consistent with the prediction model ASTM 900–02, which does not directly include the influence of the chemical elements P in PMIE [6].

Fig. 8 shows the relationship between the predicted ΔRT_{NDT} and irradiation temperature. It is indicated that a higher irradiation temperature will lead to a smaller irradiation embrittlement effect. In the range of 275–292 °C, when the irradiation temperature increases 1 °C, ΔRT_{NDT} decreases by 0.35 °C. In fact, previously Petrequin et al. [27] and Haggag et al. [28] have found the mitigative

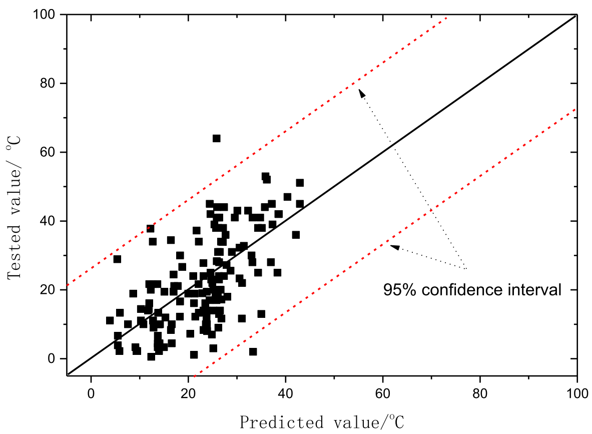

Fig. 5. The distribution of predicted values according to PMIE-2020 and tested value.

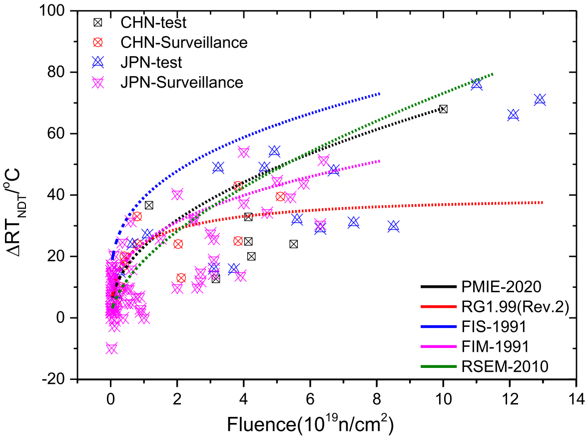

Fig. 6. The comparison between calculation results of prediction model and irradiation data for low copper alloy RPV.

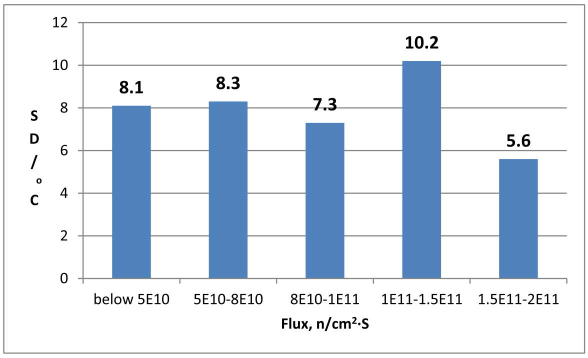

Fig. 7. The residual standard deviation of PMIE-2020 between different neutron flux.

effect on embrittlement through with neutron irradiation tests. Odette et al. further pointed out that ΔRT_{NDT} increases by 0.4–2 °C once the irradiation temperature (between 260 and 316 °C) decrease 1 °C by analyzing the irradiation data. This conclusion is basically consistent with the trend given by PMIE-2020. On the other hand, the irradiation temperature affects the embrittlement caused by Mn. It can be seen from Fig. 8 that, when the Mn content is reduced from 1.2% to 1.0%, the difference of ΔRT_{NDT} decreases from 4.0 °C (at 275 °C) to 3.5 °C (at 292 °C). With the increase of temperature during the irradiation, the atom migration ability is strengthened. Thus the interstitial atom and the vacancy type defect compound annihilation probability increases, resulting in a decrease in the defect concentration in the matrix material. This is not suitable for the formation of a stable Mn-containing complex defects [29] and consistent with the previous analysis on irradiation embrittlement mechanism.

4. Conclusion

Based on the physical mechanism analysis on RPV irradiation embrittlement, the critical threshold of Cu content of 0.072% is obtained according to a preliminary model on irradiation embrittlement. Then a low Cu RPV steel irradiation embrittlement prediction model PMIE-2020 was developed. The residual analysis indicated that the predicted results of PMIE-2020 has no tendency with influence factors such as neutron fluence, flux, irradiation temperature, chemical elements Cu, P, Mn, Ni, Si. The residual standard deviation is 10.76 °C, which is lower than the present prediction model. The distribution between predicted values of PMIE-2020 and test values are located the area near the 45° line, indicating that PMIE-2020 has high accuracy and reliability. The residual analysis shows that the neutron flux, the chemical elements P have little influence on the residual and thus do not considered in the prediction model.

Author contribution statement

Chaoliang Xu: Conceived and designed the analysis; Analyzed and interpreted the data; Wrote the paper.
Xiangbing Liu, Yuanfei Li, Wenqing Jia, Qiwei Quan : Analyzed and interpreted the data; Wrote the paper.
Wangjie Qian, Jian Yin, Xiao Jin: Contributed analysis tools or data; Wrote the paper.

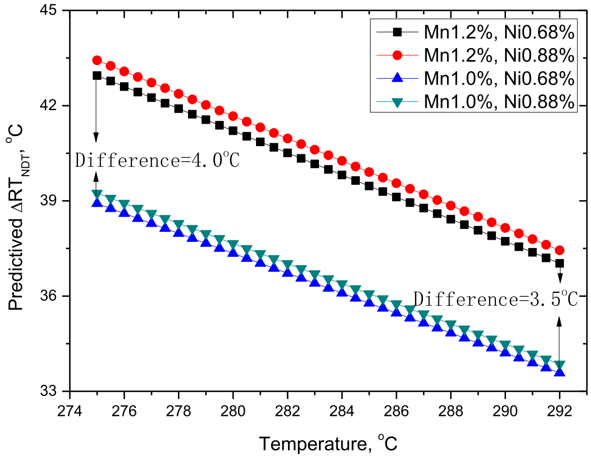

Fig. 8. The variation of ΔRT_{NDT} with irradiation temperature under different Mn and Ni content.

Data availability statement

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This research is supported by the National Key Research and Development Program of China (grant No. 2021YFA1600903), the Guangdong Major Project of Basic and Applied Basic Research (grant No. 2019B030302011), the National Natural Science Foundation of China (grant No. 12075274, U1867215, 11975171).

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