Nuclear Engineering and Technology 53 (2021) 2610–2615

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

A study of predicting irradiation-induced transition temperature shift for RPV steels with XGBoost modeling

Chaoliang Xu, Xiangbing Liu*, Hongke Wang, Yuanfei Li, Wenqing Jia, Wangjie Qian, Qiwei Quan, Huajian Zhang, Fei Xue

Suzhou Nuclear Power Research Institute, Suzhou, Jiangsu province, 215004, China

ARTICLE INFO

Article history:
Received 28 September 2020
Received in revised form
29 January 2021
Accepted 19 February 2021
Available online 26 February 2021

Keywords:
RPV
Irradiation embrittlement
XGBoost
Prediction model

ABSTRACT

The prediction of irradiation-induced transition temperature shift for RPV steels is an important method for long term operation of nuclear power plant. Based on the irradiation embrittlement data, an irradiation-induced transition temperature shift prediction model is developed with machine learning method XGBoost. Then the residual, standard deviation and predicted value vs. measured value analysis are conducted to analyze the accuracy of this model. At last, Cu content threshold and saturation values analysis, temperature dependence, Ni/Cu dependence and flux effect are given to verify the reliability. Those results show that the prediction model developed with XGBoost has high accuracy for predicting the irradiation embrittlement trend of RPV steel. The prediction results are consistent with the current understanding of RPV embrittlement mechanism.

1. Introduction

The reactor pressure vessel (RPV) is one of the most important barriers between the reactor primary circuit and the outside environment. Its integrity must be guaranteed throughout the reactor life. RPV is usually made of low-alloy steel with high strength and toughness. After irradiated by fast neutrons (E > 1 MeV) during service, a large number of atomic-scale irradiation defects and precipitates (such as Cu precipitates) will be generated. These defects and precipitations will hinder dislocation movement and cause ductility reduction, resulting in irradiation embrittlement effect. Irradiation embrittlement will lead to low-stress rupture of RPV, which will directly threaten the safe operation of nuclear power plant.

The embrittlement of RPV steels is normally monitored by corresponding surveillance program. According to surveillance program, the mechanical property changes of the test specimens irradiated in a capsule located inside the RPV are measured and the transition temperature shift (TTS) from the Charpy V-notch tests can be obtained to evaluate the current embrittlement level of RPV steels. Although TTS can be obtained through impact tests, it is not possible to evaluate irradiation embrittlement continuously and obtain the embrittlement tendency. Therefore, several irradiation embrittlement prediction models were developed according to irradiation embrittlement mechanism, including US RG1.99 (Rev. 2) [1], NUREG/CR-6551 [2] and ASTM E900-02 (Rev. 2007) [3], France RCC-M ZG3430 [4] and RSEM B7213 [5], Japan JEAC 4201 [6], etc.

Although the existing models have been improved, due to insufficient understanding of the RPV irradiation embrittlement mechanism (for example, the effect of irradiation temperature on irradiation flux for RPV steels with different Cu contents is limited; the influence of Ni/Mn/Si precipitates on embrittlement at high fluence condition is not clear; there is still no enough embrittlement understanding for RPV irradiation more than 40 or 60 years), it is difficult to obtain further development by using traditional method to improve model accuracy and expand its scope (up to 60 or even 80 years).

Machine learning (such as eXtreme Gradient Boosting, XGBoost) is a process of summarizing a large amount of known information or data and forming timely judgments, decisions and predictions when encountering new problems. Previous studies indicated that machine learning can be used to explain many changes in irradiation damage field. For example, machine learning methods can be used to predict hardening and Charpy TTS in irradiated steels [7,8]; the artificial neural network (one of the machine learning methods) is used to establish irradiation embrittlement prediction model for RPV steel [9] and predict irradiation induced change in yield stress

* Corresponding author.
E-mail address: liuxbing@cgnpc.com.cn (X. Liu).

https://doi.org/10.1016/j.net.2021.02.015

[10]. Therefore, the machine learning method can be used to make complex and subtle predictions of irradiation induced change from a relatively small database of irradiation damage results [9]. Compared with traditional prediction model development process, machine learning does not consider the specific irradiation embrittlement mechanism, but directly starts from the irradiation embrittlement data, and predicts the irradiation properties by analyzing the internal connection of the data.

In this study, based on the collection of irradiation embrittlement data, the stratified sampling method was used first to divide the data into training data set and test data set; then a machine learning method XGBoost was used to establish the irradiation embrittlement prediction model of RPV steels; finally, the model accuracy is analyzed, and its reliability is verified according to the current irradiation embrittlement mechanism.

2. Method

2.1. Irradiation embrittlement data

The prediction model of irradiation-induced TTS is based on irradiation embrittlement data. Irradiation embrittlement data determine the reliability and application scope of prediction model. This studies collected partial international embrittlement data and Chinese domestic RPV steel embrittlement data of SA508-3 and 16MND5. The data subjects include material type, chemical composition (including Cu, P, Mn, Ni, Si, etc.) and neutron fluence, flux, irradiation temperature, TTS, etc.

Fig. 1 is the TTS variations with irradiation fluence and Cu content. It is indicated that the chemical element Cu has a significant effect on irradiation embrittlement. Therefore, the chemical element Cu is a sensitive factor and can be set as the stratified sampling factor. Table 1 is category used in stratified sampling method based on the Cu content. Compared with random sampling, stratified sampling can ensure that embrittlement data with different Cu category be covered in test set and training set. This can reduce prediction errors. In this study, 390 groups of data are collected. These data are divided into training set (80%, 312) and test set (20%, 78).

2.2. XGBoost modeling

RPV irradiation embrittlement is a complex phenomenon related to many identified or unidentified effect factors. The development of RPV prediction model is a process to find correlation between these factors. XGBoost is one kinds of machine learning method used to identify the relationships between the output and input data. It is a gradient-based boosting integration algorithm, which is an improvement over the traditional Gradient Boosting (GBDT) algorithm [11]. XGBoost performs a second-order Taylor expansion on the objective function and adds a regularization term to the objective function to obtain an optimal solution. The objective function of XGBoost includes its own loss function and regularization term, as shown in formula (1):

$$\mathrm{Obj} = \sum_{i=1}^{n} l(y_i, \widehat{y}_i) + \sum_{i=1}^{K} \Omega(f_i) \tag{1}$$

In formula (1), $l(y_i, \widehat{y}_i^{(t)})$ is the loss function, that is, the deviation between the predicted value and the true value; $\Omega(f_i)$ is the regularization term, which is the complexity degree of the tree model.

In the process of XGBoost modeling, the fluence, flux and temperature parameters in irradiation embrittlement data were normalized with Bayesian function in the range 0–1 primarily due to the magnitude being higher by several orders compared to other input variables.

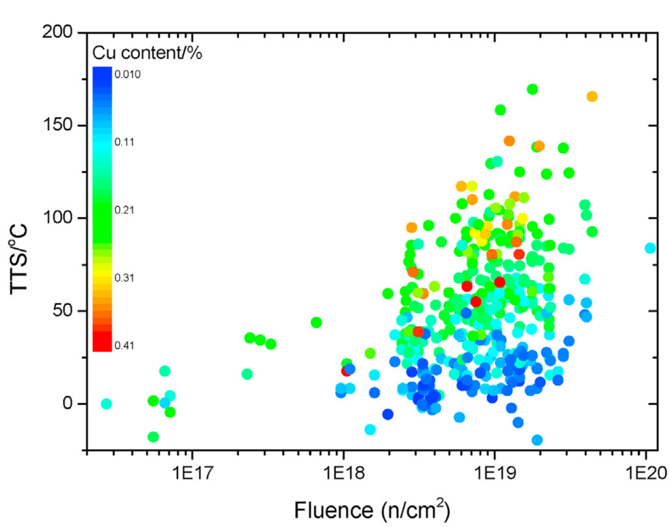

Fig. 1. The variations of TTS against the neutron fluence and Cu content.

Table 1
The category used in stratified sampling method.

Category	Cu content/wt.%
1	$0 \le Cu \le 0.07\%$
2	$0.07 \le Cu \le 0.11\%$
3	$0.11 \le Cu \le 0.21\%$
4	$0.21 \le Cu \le 0.31\%$
5	$Cu > 0.31\%$

3. Results

3.1. Model evaluation

The test data set was used to perform residual analysis for this prediction model, where the residuals refer to the difference of TTS by prediction model and Charpy impact measurement. Due to only 78 data in the test set, another 78 data are randomly selected from train set and added into test set. The least squares method was used to find the best fit by minimizing the sum of squared residuals. So a good fit will have relatively small residuals overall. Moreover, interactions of key variables can also be analyzed, and a fit that adequately reflects interaction nonlinearities will show no significant trend in the interactions. If the slope of the residual trend is significantly different from zero for a variable that is in the model, it indicates that the model does not accurately describe the effect of that variable. If the slope of the residual trend is significantly different from zero for a variable that is not in the model, it indicates that the variable should be included in the model for a better fit [12].

Fig. 2 is the correlation of residual against the irradiated fluence, flux, temperature, Cu, P, Mn and Ni. Residuals results indicate that there is a weak correlation between residual fitting curve and fluence, flux, temperature, Cu, P, Mn and Ni. The residual fitting curve is around at residual = 0 and do not show a significant trend (the maximum deviation does not exceed 3.5 °C). This indicates that the prediction model developed by XGBoost can accurately describe the correlation between TTS and irradiated fluence, flux, temperature, Cu, P, Mn and Ni.

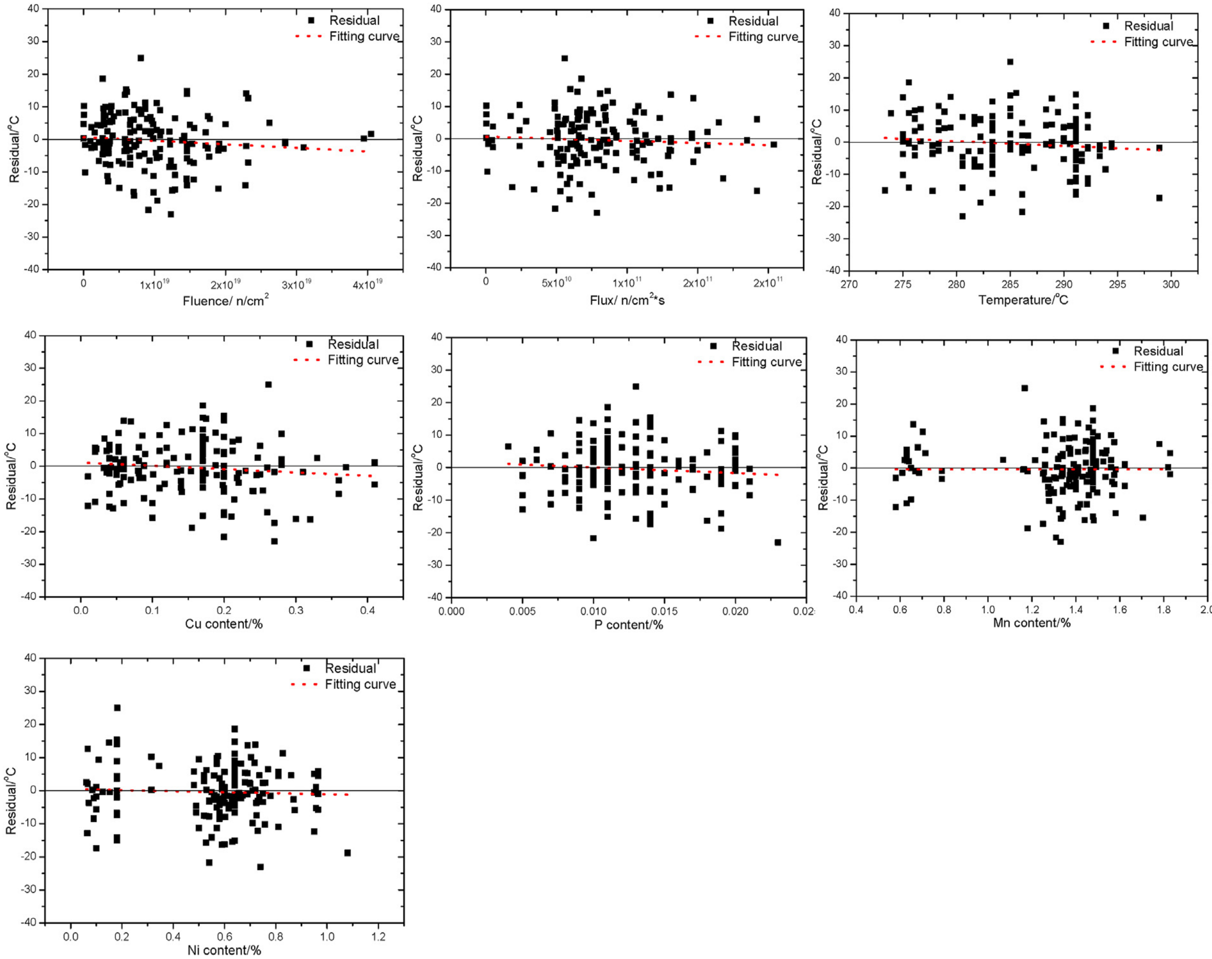

Fig. 2. The residual analysis against neutron fluence, flux, temperature, Cu, P, Mn and Ni.

A common statistical indicator of quality of fit is standard deviation (S_d) of residuals. The obtained residual standard deviation of this prediction model developed with XGBoost is 9.6 °C, which is smaller than other models according to physics mechanism (such as RG1.99 (Rev. 2) 14.8 °C, NUREG/CR-6551 12.8 °C, ASTM E900-02 12.2 °C, models developed by Mark [13] 11.9 °C, and Eason [12] 10.88 °C) as shown in Fig. 3. This indicates that prediction model developed with XGBoost has higher accuracy.

The overall assessment of fit quality can be given in the predicted TTS vs. measured TTS plots as shown in Fig. 4. It can be seen that the predicted TTS has a good consistency with the measured data. Most of the data fall in the vicinity of the 45° line, and the

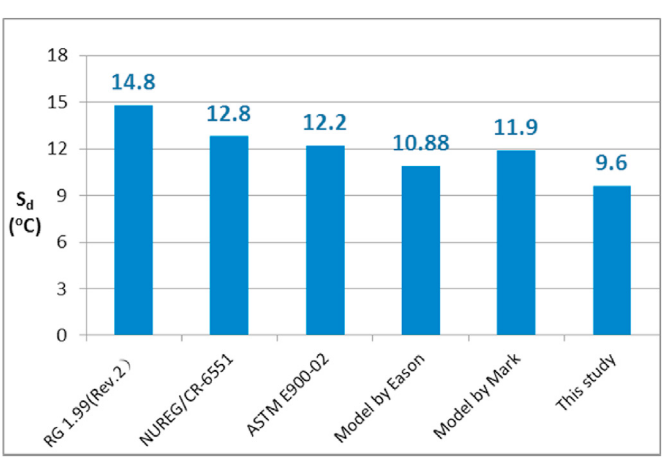

Fig. 3. The comparison of standard deviation of residual for RPV prediction models.

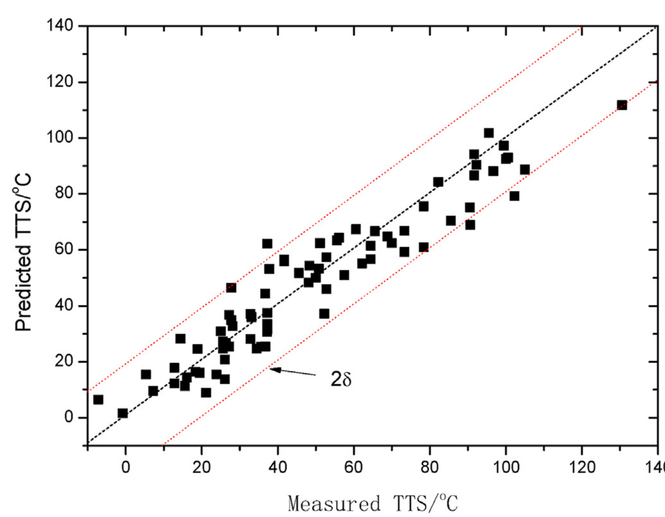

Fig. 4. The distribution of predicted TTS vs. measured TTS.

distribution is relatively uniform. Most of the data is basically within the 95% confidence interval, indicating that the current model developed with XGBoost has high accuracy for predicting the irradiation embrittlement trend of RPV steel.

3.2. Model analysis

3.2.1. Cu content threshold and saturation value

Previous studies [14] have shown that, there is a threshold value for the chemical element Cu on irradiation embrittlement. Below this threshold value, the chemical element Cu has no effect on irradiation embrittlement. According to the physical mechanism of irradiation embrittlement [15], there is a relationship between TTS of low Cu RPV steel and neutron fluence and irradiation temperature:

$$TTS = A_0(1-0.002445T)f^{0.5}$$

where T is the irradiation temperature, f is the neutron irradiation fluence, A_0 is the coefficient. In order to determine the threshold value of Cu content on irradiated embrittlement, the data of Cu content less than 0.13% were grouped. Thus the relationship between the parameter A_0 and the average residual with the Cu content can be obtained as shown in Fig. 5. It is indicated that, when Cu $\leq$ 0.07%, the parameters A_0 and average residual do not change significantly with the increase of Cu content. When Cu>0.07%, the parameters A_0 and average residual show a closed relationship with Cu content. Therefore, from the point of view of irradiation embrittlement mechanism, the threshold value of Cu content on embrittlement is 0.07%; when Cu $\leq$ 0.07%, the embrittlement of RPV steel has nothing to do with Cu content. This Cu content threshold is consistent with previous studies [12]. In this study, the variation of TTS calculated from prediction model developed with XGBoost vs. Cu content is shown in Fig. 6. It is indicated that the predicted TTS is not related to the change of Cu content before Cu $\leq$ 0.07%. When Cu > 0.07% (not beyond 0.26%), the predicted TTS increases gradually with the increase of Cu content. This indicates that the threshold value of Cu content obtained from predicted model developed with XGBoost is consistent with that obtained from irradiation embrittlement mechanism.

On the other hand, the effect of Cu content on irradiation hardening and embrittlement has a maximum value (or saturation value). If Cu content exceeds this saturation value, Cu has no effect on irradiation embrittlement. At present, there are different understandings on Cu saturation value in prediction models developed with irradiation embrittlement physical mechanisms. For example, the Cu saturation value determined by NUREG/CR-6551 is 0.30%, by ASTM E900-02 (Rev. 2007) is 0.25% (for Linde 80 and Linde 0091 weld) and 0.305% (for other materials), by EONY model [12] is 0.243% (0.5%≤Ni ≤ 0.75%). In present study, as shown in Fig. 6, the Cu saturation value obtained from prediction model developed with XGBoost is about 0.26%, which is basically consistent with the current understanding of prediction model based on physical mechanisms.

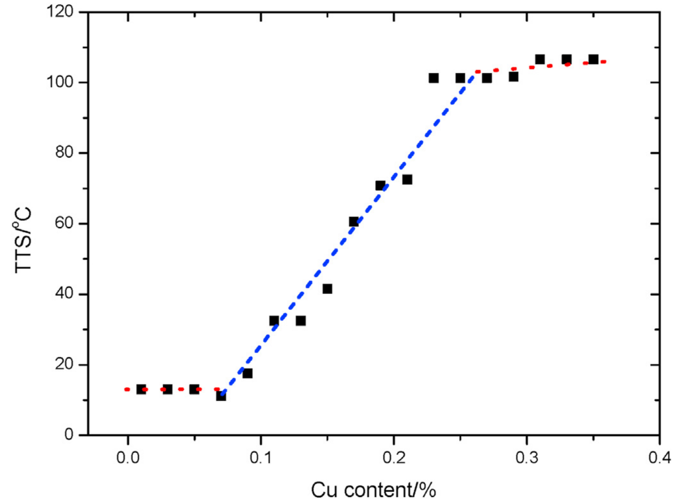

Fig. 6. The variations of TTS calculated from model developed with XGBoost vs. Cu content (input parameter: fluence: 1 × 10^{19}n/cm^2; T:290 °C; Mn:1.4%; P: 0.01%; Si: 0.25%; Ni:0.7%).

3.2.2. Temperature dependence

Temperature has an important effect on irradiation damage due to the effect on the balance between defect creation, migration and annihilation. A large number of studies have shown that, there is a negative correlation between irradiation embrittlement and irradiation temperature, that is, the higher the temperature, the smaller the irradiation effect [16]. This is because, during the irradiation process, with the increase of temperature, the atom migration ability is strengthened; the probability of annihilation of interstitial atoms and vacant defects increases, resulting in a decrease in the defect concentration in the matrix material. This is not beneficial to the formation of stable complex defects.

Fig. 7 shows the TTS under different temperature conditions according model developed with XGBoost. Overall, irradiation embrittlement is gradually obvious as the temperature decreases with an approximate linear dependence. This is in good agreement with the current understanding of irradiation damage [17] as discussed above and the negative correlation TTS∝1.869−0.00457T proposed by Jones et al. [15].

Previous prediction models, such as NUREG/CR 6551 [2] and

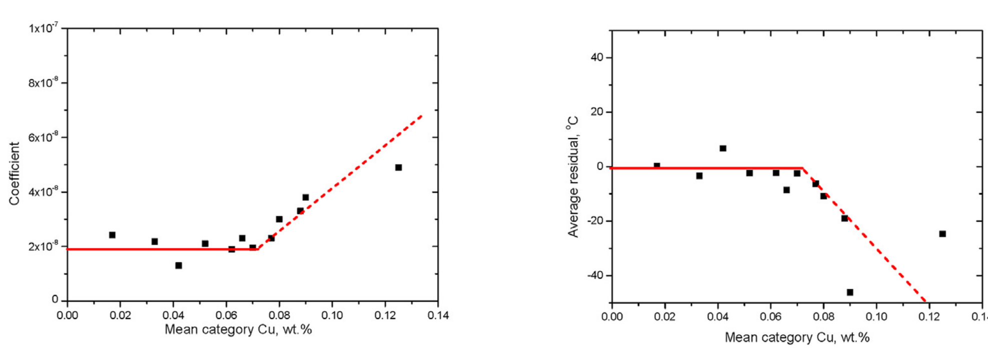

Fig. 5. The variations of coefficient A_0 and average residual vs. Cu content.

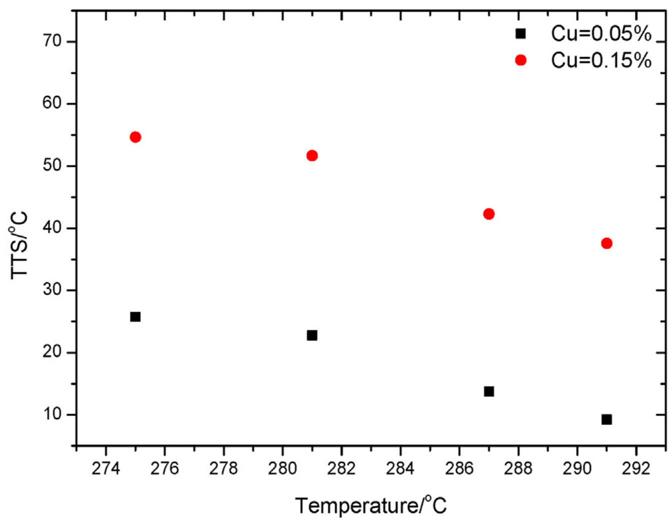

Fig. 7. TTS with irradiation temperature according to model developed with XGBoost.

ASTM E900-02 (Rev. 2007) [3], indicated that the influence of irradiation temperature effect is only related to the matrix damage. Whereas in our studies, the TTS increment is about 1.0 °C per degree temperature decrease for Cu = 0.05% steel and 1.1 °C per degree temperature decrease for Cu = 0.15% steel. A different TTS reducion exist in RPV steels with different Cu contents. This indicates that the influence of irradiation temperature on embrittlement is not only related to the matrix damage, but also related to the Cu content. This understanding is consistent with the results of ENOY model [12] and may be due to more Cu precipitates fromed at lower temperature. More studies are needed in future.

3.2.3. Ni/Cu dependence

The Ni/Cu dependences of irradiation embrittlement are important because the RPV embrittlement is sensitive to the Ni and Cu content. Fig. 8 shows the TTS against fluence for RPV steels with different Cu and Ni contents. It is indicated that more significant irradiation embrittlement phenomenon can be observed in higher Cu or higher Ni steel. On the other hand, from Fig. 8, an average TTS caused by Cu increase (from 0.05% to 0.15%, Ni = 0.1%, 1×10^{19} to 4.5×10^{19} n/cm^2) is 24.9 °C and by Ni increase (from 0.1% to 0.7%, Cu = 0.05%, 1×10^{19} to 4.5×10^{19} n/cm^2) is 5.3 °C. That is to say, the effect of Ni and Cu on TTS increment separately is 5.3 °C and 24.9 with present parameters and thus causes a total TTS increment 30.2 °C. While, a total TTS increment is 35.9 °C under Cu = 0.15% and Ni = 0.7% conditions. This indicates that Cu and Ni not only increase the embrittlement of RPV steels separately, but also have a synergistic effect. Previously, small-angle neutron scattering (SANS) results [18] indicate that, in the A533B sample, as the Ni concentration increases, the average size of copper-rich atom clusters decreases and the cluster density increases. Atom probe topography (APT) analysis of Cu-rich clusters shows that the precipitation of Cu is accompanied by alloying elements such as Ni. The precipitated clusters of Ni often become the nucleation sites for the precipitation of Cu elements. Therefore, the effect of Ni on irradiation embrittlement is caused by the contribution to the Cu-rich precipitation. So a remarkable increase will be observed in high Cu and High Ni steel.

3.2.4. Flux dependence

The dependence of irradiation embrittlement on flux is a very important question because sometimes the accelerated irradiations with high flux in experimental reactor is needed in order to reach a required fluence as soon as possible. However, the understanding of flux dependence on irradiation embrittlement is inconsistent at present [19]. The traditional view is that, interstitial and vacancy pairs with high flux irradiation will increase matrix recombination rate which results in a shorter average lifetime and therefore a lower overall mobility of solute atoms. So high flux irradiation may cause less total embrittlement. However, the understanding above does not consider the complex chemical composition and service temperature. In the case of RPV steels, whether this understanding is correct or not need more studies.

In order to confirm flux dependence or independence, the variations of irradiation embrittlement under different neutron flux according to prediction model developed with XGBoost are shown in Fig. 9. It is indicated that, although a small decrease of TTS with increase the flux, considering the error range (the maximum residual is about 2 °C according to the residual analysis), no significant flux effect on irradiation embrittlement can be observed up to 5×10^{11} n/cm^2·s. In fact, previous studies indicated that no flux dependence can be observed in RPV steels below 5×10^{11}n/cm^2·s [20]. This result is consistent with our present studies.

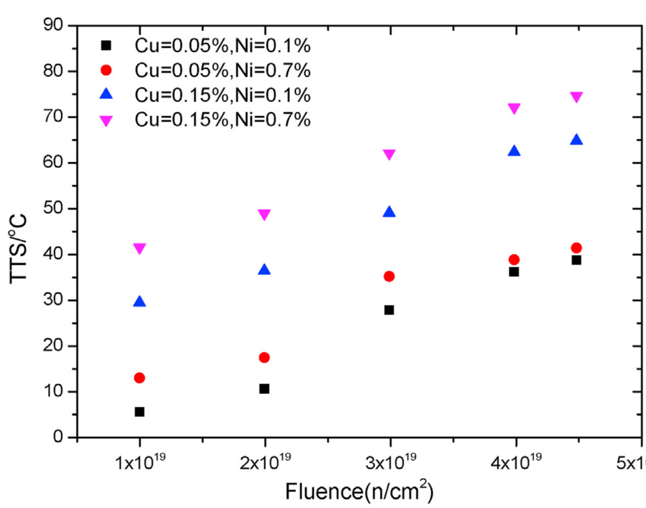

Fig. 8. The TTS with the neutron fluence under different Cu and Ni content according to model developed with XGBoost.

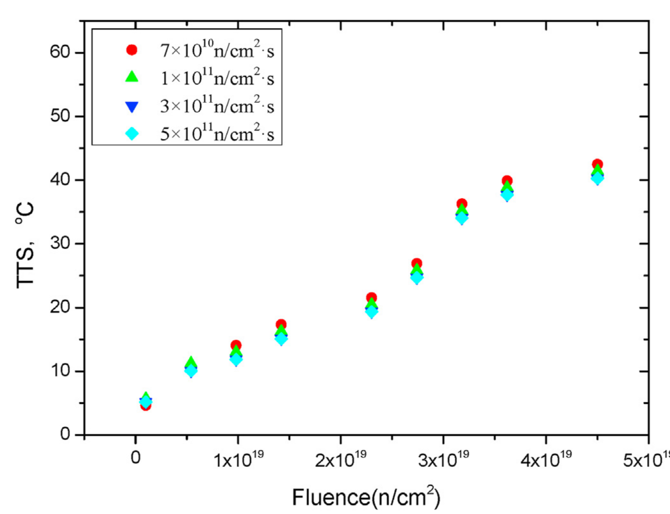

Fig. 9. The TTS under different neutron flux according to model developed with XGBoost.

4. Conclusions

The prediction of irradiation-induced TTS for RPV steels is an important method for long term operation of nuclear power plant. Based on the irradiation embrittlement data, an irradiation-induced transition temperature shift prediction model is developed with machine learning method XGBoost. Then the residual, standard deviation and predicted value vs. measured value analysis are conducted to analyze the accuracy of this model. At last, the analysis on Cu content threshold and saturation value, temperature dependence, Ni/Cu dependence and flux dependence are given. The analysis results show that the prediction model developed with XGBoost has high accuracy for predicting the irradiation embrittlement trend of RPV steel. The prediction results are consistent with the current understanding of the embrittlement mechanism of RPV.

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. 2017YFB0702204) and Guangdong Major Project of Basic and Applied Basic Research (grant No. 2019B030302011) and the National Natural Science Foundation of China (grant No. 12075274).

References

[1] 99 l Regulatory Guide, Radiation Embrittlement of Reactor Vessel Materials (Revision 2), Nuclear Regulation Commission, USA, 1988.
[2] Improved Embrittlement Correlations for Reactor Pressure Vessel Steels, NUREG/CR-6551, Nuclear Regulatory Commission, USA, 1998.
[3] ASTM E 900-02, Standard Guide for Predicting Radiation-Induced Transition Temperature Shift in Reactor Vessel Materials, 2007.
[4] RCC-M, Design and Construction Rules for Mechanical Components of PWR Nuclear Ialands, AFCEN, France, 1993.
[5] RSE-M (Addendum 2005), In-Service Inspection Rules for the Mechanical Components of PWR Nuclear Islands, AFCEN, France, 2005.
[6] Nuclear Reactor Pressure Vessel Structural Material Surveillance Test Method, JEAC 4201, JEAC, Japan, 1991.
[7] G.A. Cottrell, R. Kemp, H.K.D.H. Bhadeshia, et al., J. Nucl. Mater. 367–370 (2007) 603–609.
[8] R. Kemp, G.A. Cottrell, H.K.D.H. Bhadeshia, et al., J. Nucl. Mater. 348 (2006) 311–328.
[9] J. Mathew, D. Parfitt, K. Wilford, et al., J. Nucl. Mater. 502 (2018) 311–322.
[10] N. Castin, L. Malerba, R. Chaouadi, J. Nucl. Mater. 408 (2011) 30–39.
[11] T. Chen, T. He, M. Benesty, Xgboost: Extreme Gradient Boosting, 2015, pp. 1–4. R Package Version 0.4-2.
[12] E.D. Eason, G.R. Odette, R.K. Nanstad, T. Yamamoto, J. Nucl. Mater. 433 (2013) 240–254.
[13] Mark Kirk, Cayetano Santos, Ernest Eason, Joyce Wright, G. Robert Odette. Updated embrittlement trend curve for reactor pressure vessel steels[C]// Transactions of the 17th International Conference on Structural Mechanics in Reactor Technology (SMiRT 17) Prague, Czech Republic, August 17-22, 2003.
[14] G.R. Odette, G.E. Lucas, Irradiation embrittlement of reactor pressure vessel steels: mechanisms, models, and data correlation, in: Radiation Embrittlement of Nuclear Reactor Pressure Vessel Steels: an International Review (Second Volume), ASTM STP 909, 1986, pp. 206–241.
[15] R.B. Jones, T.J. Williams, The dependence of radiation hardening and embrittlement on irradiation temperature, in: Effects of Radiation on Materials: 17th International Symposium, ASTM STP 1270, 1996, pp. 569–590.
[16] F.M. Haggag, Effects of irradiation temperature on embrittlement of nuclear pressure vessel steels, in: Effects of Radiation on Materials: Sixteenth International Symposium. ASTM STP 1175, 1994, pp. 172–185.
[17] L. Debarberis, B. Acosta, A. Zeman, et al., Scripta Mater. 53 (2005) 769–773.
[18] D.R. Harries, Annual Report on Underlying Research at Harwell, vol. 15, 1983.
[19] A. Ballesteros, R. Ahlstrand, C. Bruynooghe, et al., Prog. Nucl. Energy 53 (2011) 756–759.
[20] G.R. Odette, T. Yamamoto, D. Klingensmith, On the effect of dose rate on irradiation hardening of RPV steels, Phil. Mag. 85 (2005) 779–797.