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Neural network analysis of Charpy transition temperature of irradiated low-activation martensitic steels

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Abstract

We have constructed a Bayesian neural network model that predicts the change, due to neutron irradiation, of the Charpy ductile-brittle transition temperature (ΔDBTT) of low-activation martensitic steels given a set of multi-dimensional published data with doses <100 displacements per atom (dpa). Results show the high significance of irradiation temperature and $(dpa)^{1/2}$ in determining ΔDBTT. Sparse data regions were identified by the size of the modelling uncertainties, indicating areas where further experimental data are needed. The method has promise for selecting and ranking experiments on future irradiation materials test facilities.

1. Introduction

Ideally, one would like to predict the behaviour of radiation-damaged materials in future fusion power plants using mechanistic models [1], allied with the experimental data from past and present facilities as well as future ones (*e.g.* the International Fusion Materials Irradiation Facility (IFMIF) [2]). The fusion materials problem is an example of a complex technology where the science is not fully established because of the large number of variables that determine properties, and where ignoring any of these variables could lead to a loss of vital information [3]. The fracture toughness of irradiated steel is currently impossible to predict given a detailed description of the chemical composition, heat treatment, neutron irradiation parameters, irradiation temperature *etc.* Commonly used methods for empirical data fitting are limited by the need to specify *a priori* fitting functions and are therefore incapable of capturing unforeseen non-linear couplings between variables, thus limiting their validity when making predictions outside the data range of the models.

In treating data, there are two kinds of errors to consider. *Noise* is the familiar scatter which results when an experiment is repeated. It arises because there are variables which are not controlled. *Uncertainty* arises because there may exist many mathematical functions which adequately represent a set of experimental data, but which behave differently

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in extrapolation. This *modelling uncertainty* becomes large when data are sparse or badly scattered and is of prime importance when dealing with non-linear fitting because it highlights the problems of extrapolation and interpolation.

This paper describes development of a neural network (NN) model for the ductile-brittle transition temperature shift (ΔDBTT) in irradiated low-activation martensitic (LAM) steels. We have previously modelled the tensile properties of such steels using a NN [4]. Toughness is related to the ability of a material to absorb energy during fracture. The Charpy test involves the measurement of the energy required to fracture a square section notched bar. Ferritic steels have a DBTT in which the fracture mode changes from ductile at high temperatures to brittle at low temperatures, and this can be measured by conducting Charpy tests at different temperatures.

2. The database and network design

Data on the impact properties of LAM steels were compiled from the literature [5], including leading candidate alloys such as F82H (8Cr) and Eurofer 97 (9Cr) as well as the conventional T91 (9Cr–1Mo), EM10 (9Cr–1Mo) and 2.25Cr bainitic grades. This resulted in a total of about 450 experimental sets for the ΔDBTT data with the parameters and ranges listed in Table 1. The inputs include: chemical composition, a parameter to describe the coldwork of the steel, heat treatment data, neutron irradiation dose ϕ_{irr} (dpa) and irradiation temperature T_{irr}. Some missing data prevented us from including the following inputs: dose rate, helium content, tensile yield stress and fracture mode. Some iron specimens were included, for which heat-treatment temperatures were set to zero °C. To avoid this causing complications, we also included a kinetic time term ($\exp(-Q/kT)$) – where Q is an activation energy for defect motion – which is zero for no heat treatment time. The activation energy, Q, for self-diffusion in ferritic iron is 240 kJ per mole. For un-irradiated specimens in the dataset, ΔDBTT was set to zero. We have restricted the data to sub-sized Charpy specimens with cross sections ranging from 3.3×3.3 mm to 3×4 mm to avoid possible size-complication effects [6].

Details of neural networks have been given by numerous authors and relevant descriptions of applications in the materials science field appear in [3]. In summary, a typical NN consists of an input layer (in this work with 35 nodes, corresponding to the 'raw' inputs in Table 1 plus nine additional inputs derived from these, such as $(\text{dpa})^{1/2}$), a 'hidden' layer in which the number of nodes (typically 2–15 nodes) determines the complexity of the model, and an output layer giving ΔDBTT. Although the network can theoretically infer functional relationships such as $(\text{dpa})^{1/2}$ or kinetic time, including such inputs as well as the unmodified data allows simpler models to be created if such inputs explain the data better than the unmodified inputs. If these inputs do not explain the data well, the NN will effectively ignore them. This approach allows different model physics to be compared, without biasing the model.

Table 1
Variables and their statistics used in developing the ΔDBTT model

Input	Data range	Mean ± s.d.
Normalising temperature (°C)	0–1100	1024.6 ± 95.7
Normalising time (h)	0–2	0.67 ± 0.33
Tempering temperature (°C)	0–780	740.2 ± 83.0
Tempering time (h)	0–2.5	1.71 ± 0.502
Coldwork (%)	0–27	0.941 ± 4.958
C (wt%)	0.005–0.2	0.115 ± 0.025
Cr (wt%)	0–12	8.85 ± 1.951
W (wt%)	0–2.12	1.009 ± 0.889
Mo (wt%)	0–1	0.207 ± 0.340
Ta (wt%)	0–0.48	0.033 ± 0.046
V (wt%)	0–0.314	0.218 ± 0.064
Si (wt%)	0–0.4	0.173 ± 0.123
Mn (wt%)	0.04–1.35	0.496 ± 0.269
N (wt%)	0.0007–0.06	0.018 ± 0.012
Al (wt%)	0.001–0.054	0.015 ± 0.015
B (wt%)	0–0.009	0.003 ± 0.003
Co (wt%)	0.0024–0.02	0.006 ± 0.004
Cu (wt%)	0.0017–0.035	0.012 ± 0.011
Nb (wt%)	0.0001–0.2	0.035 ± 0.065
Ni (wt%)	0.005–2	0.181 ± 0.332
P (wt%)	0.002–0.016	0.007 ± 0.005
S (wt%)	0.0002–0.008	0.004 ± 0.002
Ti (wt%)	0.001	0.006 ± 0.003
Zr (wt%)	0–0.059	0.007 ± 0.017
Irradiation temperature (°C)	60–550	352.7 ± 71.041
Irradiation dose (dpa)	0–100	5.42 ± 10.21
Range of ΔDBTT (°C)	(−)39–335	67.21 ± 63.66

The dataset was randomly partitioned into equal-sized *training* and *test* sets. The network is created using the training data and its ability to generalize assessed using the test data. Excessively simple or complicated models lead to large errors in predicting the test data set. The level of model complexity necessary to best fit the available data corresponds to a minimum in this *test error*, penalising both under- and over-complex models. The performance

was further improved by making predictions using a committee of different neural networks [3,4].

Unlike conventional regression, the training does not simply find a 'best fit' set of network weight coefficients, but instead uses a Bayesian inference method [7] to assign a probability distribution of weights to the network. This approach has the advantage that the parameters describing the distribution give a measure of the modelling uncertainty. This quantity represents the spread of different functions which may fit the data, with the model prediction being the most likely value. It is this feature which allows reliable models to be trained from small datasets – a small number of data will have many possible fitting functions (for a given model complexity) and hence large uncertainties. Increasing the data available will reduce the number of possible fitting functions and hence the associated modelling uncertainty. There are examples in the literature of predictions made, using this method, which had large uncertainties due to being far from the training dataset, but which ultimately proved to be very close to experimental values [8].

3. Results

3.1. Model training and performance

To illustrate the ability of the model to fit the data, Fig. 1 shows the performance of the best 14-member committee to predict ΔDBTT for the entire dataset. There are very few *outliers, i.e.*, points whose $\pm 1\ \sigma$ modelling uncertainties do not intersect the line of unit slope.

3.2. Significance of individual inputs

The significance of each input in contributing to the output (Fig. 2) is estimated from the variance of the network σ_w, and is roughly equivalent to a partial correlation coefficient [7]. The normalizing and tempering temperatures and times are found to be significant inputs in predicting ΔDBTT. ΔDBTT is more strongly correlated with T_{irr} than simply with dose. This result suggests that thermal mobilization of irradiation defects plays a greater role than their absolute density, but the trend is also

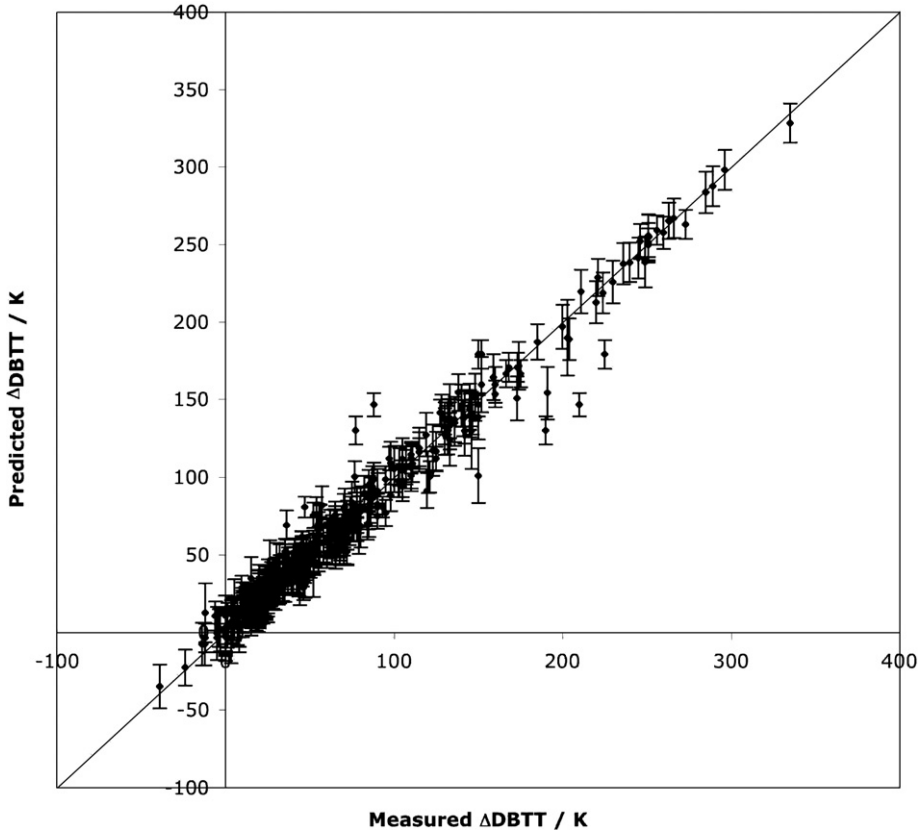

Fig. 1. Model-predicted change in DBTT (ΔDBTT after irradiation) versus the measured value for all the points.

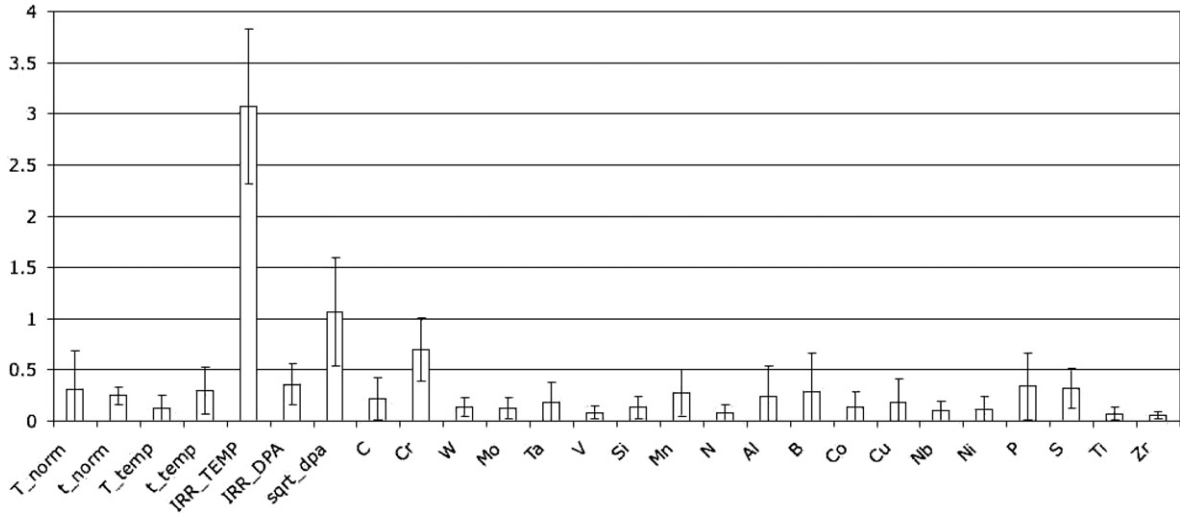

Fig. 2. Perceived model significances for ΔDBTT of heat treatment parameters, irradiation parameters and chemical element composition. The histograms represent the mean significance of all 14 committee members for each parameter and the error bars the standard deviation within each group. Some low-significance input parameters have been excluded (*e.g.* coldwork) to make the chart easier to read.

consistent with the possible operation of irradiation damage recovery processes mitigating the ΔDBTT shift for higher irradiation temperatures. The significance of the physically-motivated function $(\text{dpa})^{1/2}$ is larger than for dpa alone. A function of this form is consistent with models [5] either for solute depletion in a precipitation hardening mechanism, or with an excluded volume effect for the accumulation of knock-on damage defects.

The Cr concentration is perceived to be important by all committee members. The addition of Cr alone provides very little solid solution strengthening, but C and Cr together promote carbide formation; it is well known that both elements determine the sequence of carbide precipitation reactions [9]. Notice that in the case of Al, B, Co, Ta, Nb, Ni and P, there is some lack of consensus between the members of the committee, indicating less reliable conclusions with these inputs.

3.3. Model predictions

There are many possible ways of using the model to make predictions. In this section, we discuss some of these.

3.3.1. Effect of irradiation temperature

The ΔDBTT model shows that T_{irr} is the input parameter with the highest significance. Fig. 3 shows its effect on ΔDBTT where the experimental points are shown to compare well with the model prediction (lines) for T91 and HT-9 (12Cr–1Mo–VW) steels with ϕ_{irr} of 13 and 26 dpa. The overall trend is a decrease of ΔDBTT with T_{irr}. This is expected since recovery processes operate more readily at high temperatures. This is also consistent with the fact that ΔDBTT reaches an asymptotic limit with T_{irr}, once the competing processes of recovery and defect creation become steady. The C concentration in HT-9 is 0.2 wt%, twice the value of that in the T91 (0.09 wt%), which could possibly explain why the asymptotic limit is higher for HT-9 than T91.

3.3.2. Effect of irradiation dose

Fig. 4 shows the effect of irradiation dose on ΔDBTT for Eurofer for three values of the T_{irr}. At the lowest $T_{\text{irr}} = 250$ °C, the ΔDBTT increases monotonically with dose. However, at the higher T_{irr} of 350 °C, the data are consistent with a possible recovery: the slope decreases and at a higher T_{irr} of 450 °C the ΔDBTT curve shows possible saturation and maximum shift of 100 °C at ~20 dpa, with a decrease at higher doses. However, the uncertainties are large, and further experiments are needed to confirm this effect.

3.3.3. Effect of chromium concentration

The variation of ΔDBTT with Cr concentration is shown in Fig. 5 for three values of T_{irr}. A clear minimum occurs at a concentration of ~9 wt%, consistent with the literature [10, p. 151]. The database

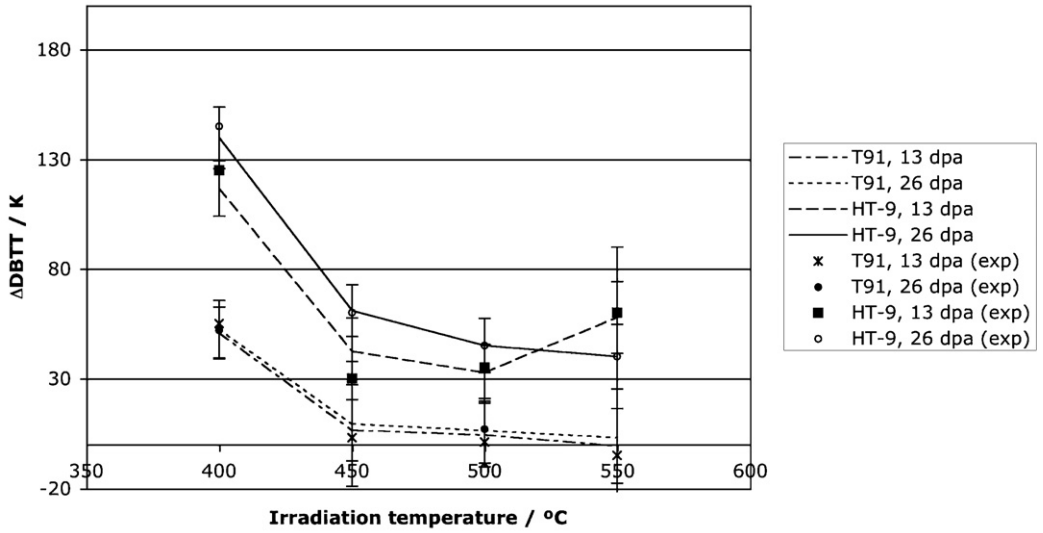

Fig. 3. Variation of ΔDBTT with irradiation temperature, comparing model predictions (lines and error bars) with experimental data (points) for T91 and HT-9 steels at 13 and 26 dpa [10,p. 141]. These data were included in the training database.

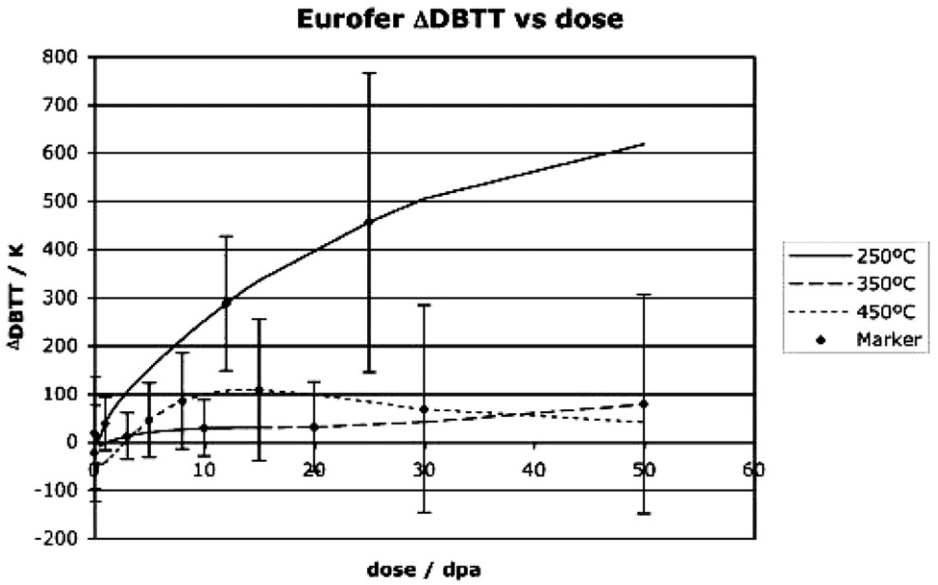

Fig. 4. Model prediction: variation (with modelling uncertainties – the markers indicate which uncertainties correspond to which line) of ΔDBTT with the irradiation dose for Eurofer for three irradiation temperatures.

has been checked to see whether ΔDBTT is linked to the un-irradiated DBTT, but no correlation was found. The result is important in that it suggests there is an optimum chromium concentration to minimise embrittlement. There is evidence [11] that for the higher Cr concentrations (~12 wt%), hardening occurs via the formation of fine-scale Cr-rich α' precipitates. The hardening will give rise to $\Delta DBTT > 0$. The situation is less clear for low Cr concentrations.

3.3.4. Effect of tantalum concentration

Tantalum was originally thought to produce carbide precipitates and thus confer strengthening in steels. However most of the Ta remains in solution after normalizing [12], and the Ta is believed to

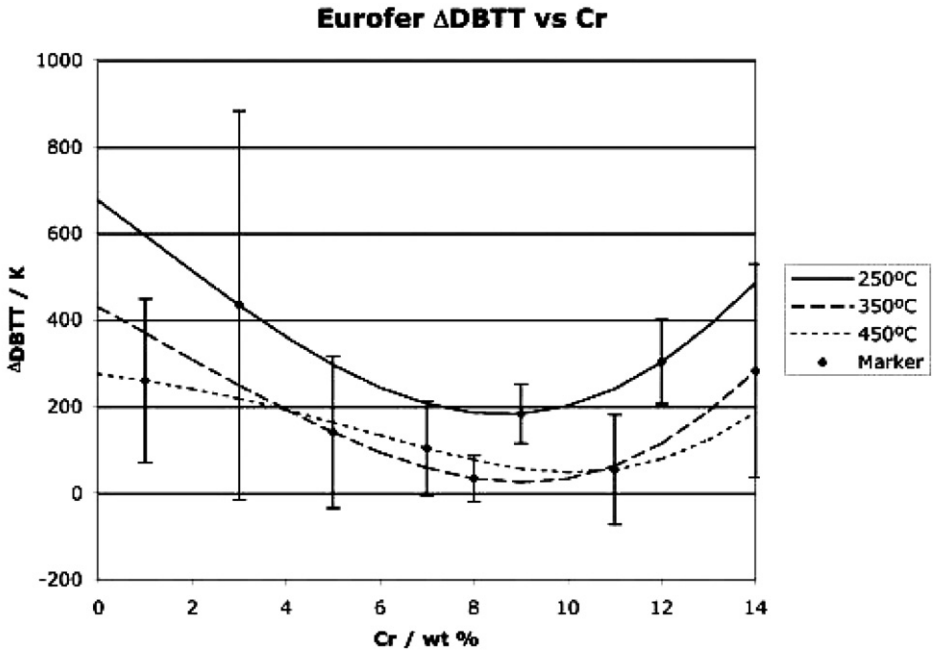

Fig. 5. Model prediction: variation (with modelling uncertainties – the markers indicate which uncertainties correspond to which line) of ΔDBTT with the Cr concentration for Eurofer for three irradiation temperatures and a dose of 10 dpa.

promote austenite grain refinement, thus improving fracture toughness. Although there was no strong NN committee consensus as to the significance of Ta concentration, we have nevertheless made a prediction. Fig. 6 shows the predicted ΔDBTT, for three values of T_{irr}. The trend is towards lower ΔDBTT at higher Ta concentrations at all T_{irr}; but the size of the error bars indicate the need for further experiments.

4. Summary and conclusions

A neural network model has been created to estimate the ΔDBTT for a database of RAFM steels,

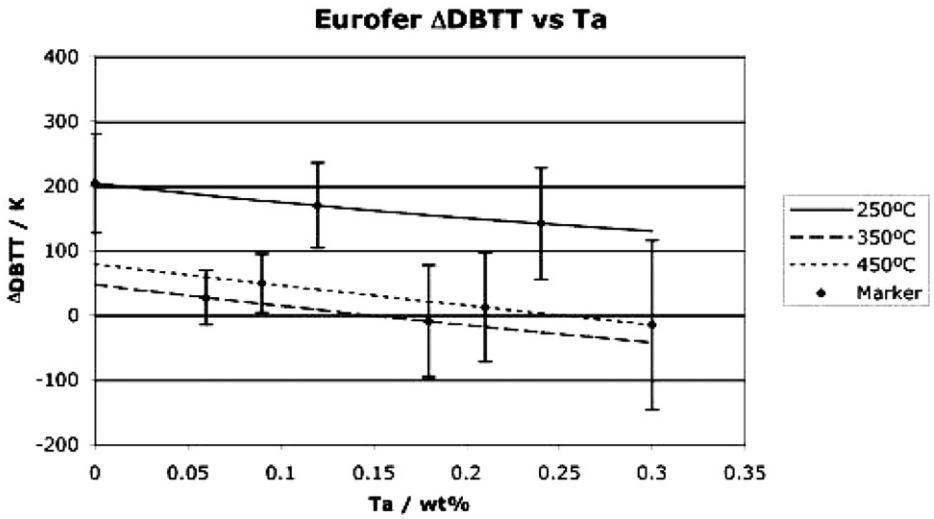

Fig. 6. Model prediction: variation (with modelling uncertainties – the markers indicate which uncertainties correspond to which line), of ΔDBTT on the Tantalum concentration for Eurofer for three irradiation temperatures and a dose of 10 dpa.

accepting a 35-dimensional input vector of material and irradiation parameters. The model needs no *a priori* fitting function, although physically-motivated functions can be included and will be selected for by the network if they are useful in explaining the data, and rejected if they are not. The model not only reproduces some well-established relationships but has also revealed some trends and features. These include:

- The ability to identify significant input parameters controlling ΔDBTT. The input with the highest significance is T_{irr} and the physically-based function $(dpa)^{1/2}$ is also important. Chemical composition (*e.g.* Cr as well as other elements) and pre-irradiation heat treatment effects are also significant.
- The tendency for ΔDBTT to decrease and saturate with increasing $T_{irr} > \sim 450$ °C.
- Recovery process for samples with high T_{irr} and high doses.
- A clear minimum in ΔDBTT with Cr concentration after irradiation.
- Evidence for a reduction in ΔDBTT with Ta concentration after irradiation.

The model is far from comprehensive since it is constructed from an imperfect database. We were forced to exclude some data through incompleteness, highlighting the need for authors to report experimental data as fully as possible. Nevertheless, results show the sensitivity of ΔDBTT to *all the inputs*.

Future work will cross-correlate the model with our previous model [4] for the tensile yield stress, and predict and compare ΔDBTT with observation. This procedure should expose any biases that are related to the use of sub-sized Charpy specimens. This knowledge is important since proposed test facilities, such as IFMIF, are constrained in their high flux irradiation volumes. We believe that the NN method could be of value in choosing optimal experiments in this context [13]. The strength of the NN approach is to recognize and assign large modelling uncertainties in sparsely populated regions of the data space, thus allowing proposed experiments to be ranked in terms of information content.

As new materials data are reported, it will be possible to refine the model and reduce its uncertainties. Models of this type make the very best use of published information, taking into account experimental noise, and permit extrapolation to the conditions of future fusion power plant.

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