MODELLING OF THE EFFECT OF IRRADIATION ON CHARPY IMPACT PROPERTIES OF STEELS.

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Surveillance schemes that monitor the effect of neutron irradiation on reactor pressure vessel materials employ Charpy impact specimens that are periodically withdrawn and tested over a range of temperatures. The resulting Charpy impact absorbed energy curves have been modelled by a three parameter relationship with the same functional form as the Burr distribution function. A method is presented for the evaluation of Charpy curves based on Bayesian inference and employing Markov chain Monte Carlo simulation to quantify the model parameters and the uncertainties.

INTRODUCTION

Integrity assessments of nuclear reactors with steel pressure vessels quantify the temperature margins between the operating temperature of plant at any given location and the temperature at which fully ductile condition is expected. The mechanical property changes produced in the pressure vessels during service by thermal and neutron irradiation effects have been monitored by a surveillance scheme which includes Charpy impact specimens. One of the main effects of neutron irradiation and temperature is to increase the ductile to brittle transition temperature the changes of which can be represented as temperature shifts. According to mechanistic models, these temperature shifts can be attributed to a combination of two hardening and one non-hardening process.

In the neutron irradiation surveillance schemes for nuclear reactors, the changes in the ductile to brittle transition temperature for the pressure vessel steels are inferred from Charpy impact energy data which have been measured regularly during the service life. The mechanical properties of ferritic steels in general and Charpy impact properties in particular are inherently scattered due to the heterogeneity of their microstructure. To be able to assess the trends in the Charpy impact energy data due to neutron irradiation doses and temperature, it is necessary to optimise the interpretation of the data by mathematical processing. This paper presents strategies which can be used for interpretation of the mechanical property data that are based on Markov chain Monte Carlo sampling methods.

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Use of these methods for the estimation of the rate of irradiation embrittlement from the analysis of Charpy impact energy data is illustrated and the results are presented.

MODELLING BACKGROUND

Charpy impact energy data measured as a function of test temperature follow a sigmoidal shape (1) which has been modelled by the Burr distribution function (2). This has the flexibility to accommodate changes in the shape of the curve brought about by material degradation during service, in particular neutron irradiation and temperature. The Burr distribution function, $F(T_i)$, is given by:

$$F(T_i) = [1+\exp\{-(T_i-\mu)/\xi\}]^{-\nu} \tag{1}$$

where T_i is the test temperature, μ, ξ and ν are the model parameters which define the location of the curve on the temperature axis, gradient and the shape of the curve. The values of $F(T_i)$ at $-\infty$ and $+\infty$ are 0 and 1, respectively. The measured Charpy impact energy values are related to the Burr distribution function by:

$$C_i = \theta_0+(\theta_1-\theta_0)\,F(T_i)+\varepsilon_i \tag{2}$$

where the parameters θ_0 and θ_1 represent the lower and upper shelf Charpy impact energy values. The variability of the data from specimen to specimen due to microstructural heterogeneity is represented by the random error term, ε_i, which can be adequately specified by a normal distribution with a mean of zero and variance σ^2 dependent on function $F(T_i)$, i.e.

$$\sigma = \sigma_1+\sigma_2[F(T_i)]^2[1-F(T_i)]^{\sigma_3} \tag{3}$$

where σ_1, σ_2 and σ_3 are the parameters for variance.

Modelling of Irradiation Damage.

The model for the mean Charpy impact energy based on the Burr distribution function given by equations (1) and (2) facilitates modelling of Charpy impact energy as a function of test temperature. Modelling of irradiation damage needs to consider changes in the model parameters due to neutron dose and irradiation temperature. Furthermore, the effect of dose rate on the rate of irradiation damage has to be assessed because data obtained from surveillance experiments are frequently augmented by data measured on specimens subjected to accelerated irradiation exposures. The ranges of irradiation temperatures and neutron spectra in the irradiation surveillance scheme are often restricted by exposure conditions in those locations which contain the monitoring specimens. To assess a wider range of irradiation conditions, specimens are frequently withdrawn from the operating reactors and then used to carry out re-irradiation exposures.

In the surveillance scheme considered in this paper, the data were generated for two single irradiation temperatures which cover only an eight degree temperature range. The main motivation for the re-irradiation experiments was the fact that irradiation damage assessments of the plant needed to be carried out for irradiation temperatures outside the surveillance temperature range. Thus the re-irradiation exposures were carried out over a temperature range greater than 100°C. For the purpose of statistical analysis, the data can be subdivided according to the irradiation condition into three different sets comprising:

(i) data measured on unirradiated specimens

(ii) data measured on surveillance irradiated specimens
(iii) data measured on specimens receiving initially a in reactor surveillance irradiation at a single temperature, followed by accelerated irradiation at several different temperatures within a temperature range greater than 100°C.

The models fitted to the data by statistical analysis incorporate considerations of physical mechanisms of irradiation damage and micromechanisms of fracture. Initially, it was necessary to perform some exploratory analysis to asses the dependence of the model parameters on the irradiation conditions. From these analysis, the model parameters were subdivided into three different types.
(1) Parameters that are independent of the irradiation condition and hence common to all three data sets. This set includes θ_0, ν, σ_1, σ_2 and σ_3.
(2) Parameters which are unique to a particular data set and the dependence of the parameters on neutron dose and irradiation temperature is not statistically discernible. This set includes the gradients of the curve, ξs, and the upper shelf energies θ_js ($j>0$).
(3) Models used only for μ in which the dependence on the irradiation variables was modelled as a continuous function of neutron dose, irradiation temperature and dose rate. In the Burr distribution function, the changes in μ are modelled in terms of temperature shifts in the ductile to brittle transition temperature. Thus, the models for μ have the same form as the statistical and mechanistic models for the temperature shift, ΔT. These models are considered below.

<u>Models for Temperature Shifts and μ.</u>

Changes in the ductile to brittle transition temperature are conventionally measured as the displacement in the temperature associated with the 40J absorbed energy level. Three main forms of these models were used to analyse the Charpy impact energy data.

The first of the three models, is a relationship for the dependence of ΔT_{40J} on the temperature and neutron dose, for exposures carried out at different irradiation temperatures. During irradiation, both the temperature and the dose rate remain constant. The mechanistically based model is given by the relationship:

$$\Delta T_{40J}=B+A\sqrt{D}(a+bT_{ir}) \qquad (4)$$

where B represents hardening due to copper precipitation, A is the rate of embrittlement due to both hardening and non hardening processes, a and b are constants, T_{ir} is the irradiation temperature and D is neutron dose.

For exposures involving only a change in the dose rate during the irradiation exposure, and the temperature held constant, the Charpy impact energy temperature shift can be modelled by a statistically based relationship:

$$\Delta T_{40J}=B+A_1\sqrt{D_1}(a+bT_{ir})+A_2\sqrt{D_q}(a+bT_{ir2}) \qquad (5)$$

where $D_q=(D_1+D_2)^{0.5}-\sqrt{D_1}$ and the subscripts 1 and 2 index the two different dose rates.

Finally, for two stage irradiation exposures involving a different irradiation temperature in each stage without the change of dose rate, a mechanistically based model for the temperature shift is given by the relationship:

$$\Delta T_{40J}=B+A\{(a+bT_{ir1})^2D_1+(a+bT_{ir2})D_2\}^{0.5} \qquad (6)$$

More complex relationships may be required if both the irradiation temperature and the dose rate are different in each of the two stages of irradiation.

BAYES INFERENCE

The Bayes theorem describes the relationship between the posterior $P(\Theta|C)$ and prior $P(\Theta)$ probability distributions and is given by:

$$P(\Theta|\mathbf{C}) = \frac{P(\Theta)\,L(\Theta|\mathbf{C})}{\int P(\Theta)\,L(\Theta|\mathbf{C})\,d\Theta} \tag{7}$$

where C represents the measured Charpy impact energy data and Θ is the notation for a set of model parameters, e.g. (θ_0, θ_1, μ, ξ, etc.). In this expression, $P(\Theta)$ is the information known about the model parameters prior to measuring the data, the likelihood function, $L(\Theta|C)$, represents the information about the model parameters coming from the data and in this case is defined as the likelihood of the measured Charpy impact energy data given a specific set of model parameters Θ. The integral in the denominator normalises the probability distribution to unity. In the calculation of the likelihood, the model parameters are treated as variables and the set of measured data are fixed quantities. In order to apply the Bayesian approach, therefore, multidimensional integrals need to be evaluated. Conventional numerical methods are cumbersome, but Markov chain Monte Carlo sampling is a more convenient method that computes the required integrals explicitly.

A Markov chain is a sequence of random values of sets of model parameters. The aim of the MCMC process is to generate over a period of time a Markov chain of the model parameters which mimics the posterior probability distribution. The most commonly used algorithms to construct a Markov chain are the Gibbs sampler and the Metropolis-Hastings (3). There are two types of Metropolis-Hastings algorithm. One involves sampling from a probability distribution which is centred on the current point in the Markov chain and the mean of the distribution changes with time. The variance of the probability distribution from which the samples are taken is scaled down so that only a small range of the possible values of the model parameters is within the sampling region. This algorithm gives rise to a large number of small moves within the chain. The second Metropolis-Hastings algorithm, samples from a probability distribution which is independent of the current point in the Markov chain. This gives rise to a small number of large moves. The variance of the distribution is inflated to extend the sampling region outside all possible values of the model parameters. To improve the efficiency and flexibility of sampling, the three algorithms referred to above have been combined to form a hybrid algorithm.

RESULTS

A large number of different computer runs were performed to optimise the model for the Charpy impact energy data. The most important results are considered below. Analysis of the data obtained over an irradiation temperature range of 14°C from re-irradiation experiments carried out for two different dose rates showed that the rates of embrittlement, A, are 5.32±2.48 and 5.66±4.62 for the surveillance and accelerated irradiation exposures, respectively. The difference between the mean values is smaller than their uncertainties indicating that the rate of embrittlement is unaffected by the dose rate. It is noteworthy that the uncertainty in the model parameter is greater for the second than for the first stage of irradiation. This is due to the fact that there are more results for the first stage than for the second stage of irradiation. The mean values of the model parameters for the gradient of the curves, ξ, for the unirradiated, surveillance irradiated and accelerated irradiated condition were found to be: ξ_1=37.7, ξ_2=41.3 and ξ_3=35.2, respectively. The 0.05 and 0.95 quantiles are typically 25 and 68, respectively. Although each of the three parameters is significant, the variability of these parameters with irradiation treatment shows no real trend and is subsumed within the uncertainty in their values. A clear trend with irradiation treatment was found in the values of the parameters $\theta_{i>0}$ for the upper shelf. The mean value for the unirradiated condition is 130, for the surveillance irradiated condition 75 and for the re-irradiated condition.

Since the dose rate has no effect on the rate of embrittlement, it is possible to disregard the fact that the dose rates were different in the two stages of irradiation and the effect of changing the irradiation temperature can be modelled by the relationship given by equation (6). Statistical analysis yielded an equation for the rate of embrittlement in the relationship for ΔT_{40J}: 5.01[1-0.00133(T_{ir}-190)].

The output obtained from Markov chain Monte Carlo simulations can be examined graphically to carry out various diagnostic checks, and obtain predictions of various quantities of interest in structural integrity assessments. Examples of histograms of the model parameters are presented in Figure 1. The measured Charpy impact energy data are compared in Figure 2 with the predictions for the mean, and 5 and 95% probability levels for both the mean and the distribution of the Charpy impact energy values about the mean. The predicted ΔT_{40J} temperature shifts as a function of irradiation temperature are presented in Figure 3.

CONCLUDING COMMENT

The Markov chain Monte Carlo sampling methods provide an effective analytical method for assessing the trends in the mechanical properties brought about by neutron irradiation and temperature.

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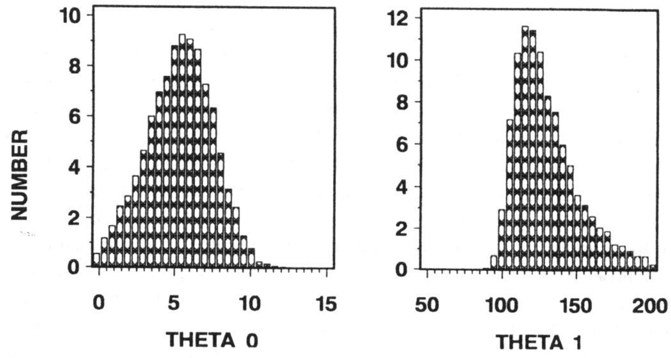

Figure 1 Histogram of model parameters for the lower, θ_0, and the upper, θ_1, shelf levels

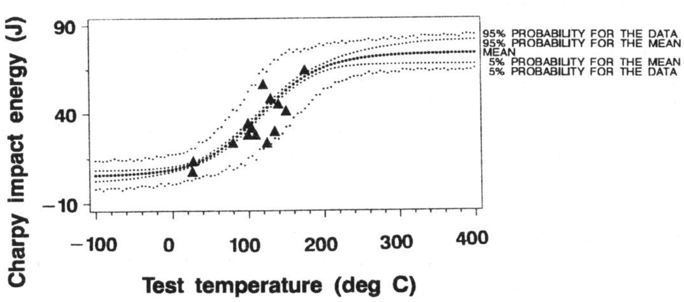

Figure 2 Comparison of fitted Charpy impact energy curves and measured data for 107.7dpa neutron dose.

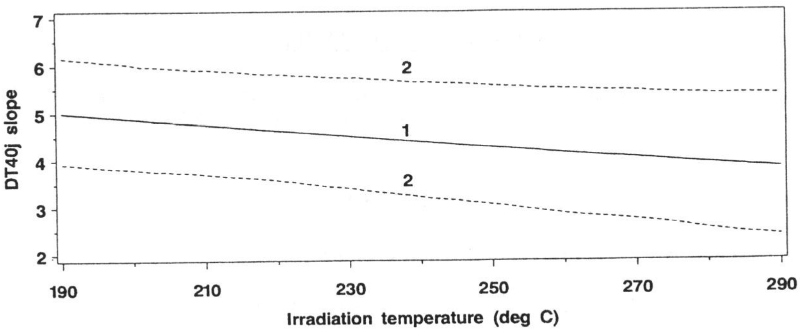

Figure 3 Predicted rate of embrittlement for ΔT_{40J} temperature shift against irradiation temperature. 1 - is the mean, 2 - are the 5 and 95% probability intervals.