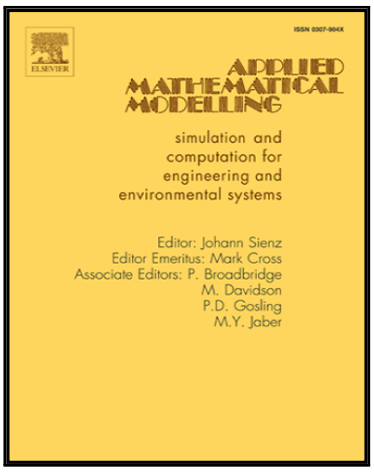

Incorporating specific batch characteristics such as chemistry, heat treatment, hardness and grain size into the Wilshire equations for safe life prediction in high temperature applications: An application to 12Cr stainless steel bars for turbine blades

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Highlights

- The Wilshire equation was modified to predict batch to batch creep properties.
- A new two step procedure was used estimate the constants of this modified equation.
- The activation energy varied between batches within the limits of 273–331 $kJmol^{-1}$.
- P,Mn,Cu, hardness and heat treatment were identified as determinants of creep life.
- The modified equation yielded safe lives more realistic to specific batches in use.

Incorporating specific batch characteristics such as chemistry, heat treatment, hardness and grain size into the Wilshire equations for safe life prediction in high temperature applications: An application to 12Cr stainless steel bars for turbine blades

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ABSTRACT

A modified version of the Wilshire equation was proposed to incorporate specific batch characteristics such as chemistry, heat treatment, hardness and grain size into the analysis of times to failure at high temperatures. A new two stage estimation procedure was proposed for obtaining values for the parameters of this modified Wilshire equation. This procedure overcomes the degrees of freedom obstacle present in extending the Wilshire equation is this direction: namely the small number of batches available in many creep data sets with which to investigate a large number of variables defining specific batch characteristics. Just a few batch characteristics were shown to be good predictors of the unknown parameters of the Wilshire equations – namely the P, Mn and Cu content of the batch, the batches hardness and some types of heat treatment. Incorporating these characteristics into the Wilshire equation produced safe life predictions which were more meaningful to specific batches of a 12Cr stainless steel alloy in that the median predictions were more representative of a particular batch data and the 0.5 - 99.5 percentile bands were much narrower and so the lower bound provided a more economically feasible safe life. The modification should allow for the more reliable safe life determination of specific batches actually being used by, for example, power generating companies.

Keywords

Wilshire equation, Time to failure, Batch characteristics, Creep

1. Introduction

The structural integrity of many high temperature components needs to be assessed at long lifetimes. In order to perform such assessments, it is necessary to have confidence in the relevant material property predictions which may involve significant extrapolation beyond the range of the available data on a given material. This can prove particularly problematic when equations are based upon polynomial expressions (such as the methods proposed by Larson-Miller [1], Orr-Dorn-Shepherd [2] and Manson-Haferd [3]), since at low stresses the equations of these models are often subject to turn-back and hence it is not possible to extrapolate to long durations. It is therefore of little surprise to note that a reduction in this 12 - 15 year materials development cycle has therefore been defined as the No.1 priority in the 2007 UK Energy Materials – Strategic Research Agenda [4].

In numerous studies on copper and various steel and titanium alloys, the Wilshire methodology [5-12] has proved remarkably successful in avoiding these extrapolation issues. However, this methodology is as yet unable to deliver accurate life time predictions for specific batches of a material because in all studies made to date using this methodology, the batch to batch variation present in large multi heat creep data sets is not modelled. That is, the methodology produces only an average or typical life time prediction for all batches of a material. As batch to batch variation is quite large, it is therefore impossible to say how long a particular batch, used within a specific power plant for example, will actually last at specified operating conditions. This would be invaluable information for power plant operators and addressing this issue forms the main content of this paper.

Batch to batch variation will contain both a systematic and a random component, especially if it is not possible to quantify all the variables that characterise in full a particular batch. Evans [13] has proposed a solution to the random nature of this batch to batch variation (within the theta methodology) and this paper extends this analysis to also include a systematic component (within the Wilshire equation). This systematic component originates from the fact that creep life is well known to depend on batch characteristics such as grain size, chemical composition, hardness and heat treatment. Alloying elements and heat treatments play a role in the formation of closely spaced precipitates that obstruct the movement of dislocations and so increase creep strength. Alloying elements play an additional role through solution strengthening, although the relative importance of all the different alloying elements is not so well understood. Details of the role played by particular alloying elements is discussed below in section 6.3.

It is also well known that grain size is important in controlling diffusional creep but not the diffusion controlled generation and movement of dislocations. Thus Burton and Greenwood [14] that in polycrystalline copper,

$$\dot{\varepsilon}_m \propto (1/d)^m$$

where d is the grain diameter and $\dot{\varepsilon}_m$ is the minimum creep rate. Burton and Greenwood found that the parameter m varied between 2 and 3 depending on the absolute temperature. There have also been a number of studies showing the importance of hardness in determining creep rupture life. Many of these studies have used the Larson-Miller equation. Thus Furtardo et. al. [15] found a strong correlation between the Larson Miller parameter and the

hardness at failure in 9Cr-1Mo ferretic steel, whilst Fujibayashi et. al. [16] found that for 2.25Cr-1Mo steel the Larson Miller parameter was a quadratic function of the log of the pre service Vickers hardness measurement, so allowing life times to be predicted from the initial hardness and stress/temperature conditions.

In principal it is possible to incorporate into the Wilshire equation specific batch characteristics such as its chemical composition, the heat treatment it has been subjected to, the grain size, the frequency of inclusions and its hardness to predict the creep properties of that particular batch. The main problem with doing this is one of estimation, in that the number of variables that define a particular batch vastly exceeds the sample size available to estimate the parameters of the Wilshire equation. As a result, standard estimation procedures, such as least squares or maximum likelihood, are inappropriate. This paper describes a two-step estimation procedure for overcoming this problem that is flexible enough in nature to deal with other life time prediction issues such as the uncertainty about the form of the failure time distribution, uncertainty about how to characterise a specific batch of a material and creep data sets that contained censored or un-failed test specimens. This new approach is illustrated using 12Cr stainless steel bar data as published by the National Institute for Materials Science (NIMS) in its creep data sheet No.13B [17].

2. The Wilshire equation

The Wilshire equation for predicting times to failure takes the form:

$$\left(\frac{\tau}{\tau_{TS}}\right) = \exp\left\{-k_1\left[t_f \exp\left(\frac{-Q_c^*}{RT}\right)\right]^{u}\right\} \tag{1a}$$

where T is the absolute temperature, τ is the stress, τ_{TS} is the tensile strength, R is the universal gas constant, Q^*_c is the activation for self-diffusion and k_1 and u are parameters requiring estimation. This can rearranged to express the log time to failure as a linear function of transformations of the normalised stress and temperature

$$\ln[t_F] = b_0 + b_1 \ln\left[-\ln\left(\tau/\tau_{TS}\right)\right] + b_2\left[\frac{1}{T}\right] \tag{1b}$$

where $b_0 = -\ln(k_1)/u$, $b_1 = 1/u$ and $b_2 = Q^*_c/R$. Applications of Eqs. (1a,b) to numerous steel and titanium alloys has revealed that the values Q^*_c, k_1 and u change is a step like fashion at some critical value for the normalised stress. One way to formulize this behavior is through the use of a spline function

$$\ln[t_f] = b_0 + b_1\tau^* + b_2\left[\frac{1}{T}\right] + b_3[\tau^* - \tau_1^*]D + b_4\left[\frac{D}{T}\right] \tag{2a}$$

where $\tau^* = \ln(-\ln(\tau/\tau_{TS}))$, τ_1^* is the value for τ^* at which b_0 , b_1 and b_4 change and D = 0 when $(\tau^*-\tau_1^*) \leq 0$ and D = 1 otherwise. This gives a two regime model where

$$\ln[t_f] = b_0 + b_1\tau^* + b_2\left[\frac{1}{T}\right] \quad ; \quad \text{when D = 0, i.e. when } (\tau^* \le \tau_1{}^*) \tag{2b}$$

but

$$\ln[t_f] = [b_0 - b_3\tau_1^*] + [b_1 + b_3]\tau^* + [b_2 + b_4]\left[\frac{1}{T}\right] \quad ; \quad \text{when D = 1, i.e. when } (\tau^* > \tau_1{}^*) \tag{2c}$$

3. The Wilshire equation modified for batch to batch variation

A potentially useful way to analyse multi batch data is to allow the parameters of Eq. (2a) to differ from batch to batch in a partly random and partly deterministic fashion. Within the lifetime statistical literature, random effects models (see Hougaard [18] for a detailed review) have been developed to account for any dependence within clusters, such as different batches of material. A hierarchical version of Eq. (2a) takes the form

$$\ln[t_f(ij)] = b_0(j) + b_1(j)\tau^*(ij) + b_2(j)\left[\frac{1}{T(ij)}\right] + b_3(j)[\tau^*(ij) - \tau_1^*(j)]D(ij) + b_4(j)\left[\frac{D(ij)}{T(ij)}\right] + \sigma(j)e(ij) \tag{3}$$

i = 1 to n(j) specimens for batch j and with j = 1 to m batches.

The letters in round brackets refer to the ith specimen cut from the jth batch of materials. Thus $t_{f(}$ij) is the failure time obtained by subjecting the ith specimen cut from the jth batch of material to a stress τ(ij) and temperature T(ij). In this formulation the parameters of the Wilshire equation differ from batch to batch as highlighted by the letter j after each such parameter. e(ij) is a standardised error term added to pick up the stochastic nature of the time at which specimens fail, i.e. the tendency for failure times to differ under identical settings of the test variables stress and temperature. The standardised error is found by dividing the actual error by the parameter σ(j), which if these errors are normally distributed would by the standard deviation of the error associated with batch j (the mean error is by definition zero).

4. A two-step estimation procedure

4.1. Step 1

In step 1, the parameters if Eq. (3) are estimated separately using maximum likelihood techniques for each batch of material. This will yield m estimates for the parameters b_0 through to b_4 and parameters τ^*_1 and σ. Let these estimates be denoted by the hat symbol as follows

$$\hat{b}_0(j) \;;\; \hat{b}_1(j) \;;\; \hat{b}_2(j) \;;\; \hat{b}_3(j) \;;\; \hat{b}_4(j); \;\; \tau_1^*(j) \;;\; \hat{\sigma}(j) \;;\; j = 1, m \tag{4}$$

One way to obtain these estimates is to apply ordinary least squares to Eq. (3) on a batch by batch basis, i.e. chose values for these parameters to as to minimise $RSS(j) = \sum_{i=1}^{n(j)} e^2(i)$ for j = 1 to m (where $\sigma^2(j)$ is further estimated as RSS(j)/n(j) - 7) that are

put on test. However this imposes some severe limitations on what can be achieved using Eqs. (3). First, because least squares does not require any specification to made about how failure times are distributed, predictions from the Wilshire equation typically come without any confidence limits placed around then. Further, least squares cannot deal with the existence of un failed or censored times, i.e. specimens within the experimental test program that have not yet failed by the time an analysis of the data is required. They must either be ignored or treated as failure times and in either approach this can lead to biased or inaccurate parameter estimates.

Using maximum likelihood is a neat alternative to least squares because not only are censored times a natural part of the estimation procedure, the need to specify a failure time distribution results in predictions being made with levels of confidence. The only concern is that the nature of the creep failure time distribution is unknown so that the best approach is to use a very general specification for the distribution. Only then is it possible to see which distribution, contained as a special case within this general specification, is actually supported by the data. One such general distribution, suggested by Bartlett and Kendall's [19], is the log gamma distribution. More recently, this distribution has been modified by Prentice [20] because in its original form the distribution had no limits. In this modification, the random variable e(ij) for each batch is taken to have the probability density function (PDF's) given by

$$f(e(ij)) = \frac{\lambda(j)^{(j)\lambda-0.5}}{\Gamma(\lambda(j))} \exp\left(\sqrt{\lambda(j)}(e(ij)/\sigma(j)) - \lambda\exp[e(ij)/(\sigma(j)\sqrt{\lambda(j)})]\right); \quad j = 1, m \qquad (5a)$$

$\Gamma(\lambda)$ is the gamma function and σ is a further parameter that standardises the random variable error. These parameters can be different for each of the j = 1, m batches and so are written as ($\sigma(j)$, $\lambda(j)$. Prentice has shown that when the parameter $\lambda(j) = 1$, e(ij) for batch j has an standard extreme value distributions (and so failure times are Weibull distributed). But when $\lambda(j) = \infty$, e(ij) for batch j has a standard normal distributions (and so failure times are log normally distributed). In this special case, $\sigma(\varphi)$ is the standard deviation for the natural log of times to failure for batch j. Further, when $\lambda(j) = 1$ and $\sigma(j) = 1$ e(ij) has an standard exponential distribution. The gamma distribution is also a special case. This specification allows for the possibility that the failure times associated with each batch can be different in nature.

Any percentile (p) of this distribution is then given by

$$\ln[t_f(ij)] = b_0(j) + b_1(j)\tau^*(ij) + b_2(j)\left[\frac{1}{T(ij)}\right] + b_3(j)[\tau^*(ij) - \tau_1^*(j)]D(ij) + b_4(j)\left[\frac{D(ij)}{T(ij)}\right] + \omega_{\lambda,p}\sigma(j) \qquad (5b)$$

j = 1, m

where

$$\omega_{\lambda,p} = \sqrt{\lambda(j)}\ln\left(\frac{1}{2\lambda(j)}\chi^2_{2\lambda,p}\right) \qquad (5c)$$

with χ^2 having a value that corresponds to the p^{th} percentile of the chi square distribution with $2\lambda(j)$ degrees of freedom.

In maximum likelihood estimation, the parameters of Eq. (5a) are chosen so as to maximise the joint probability of observing all the observed failure times and all the observed censored times recorded for batch j (where there are n(j) such observation in each batch). Such a maximisation is carried out for each batch. This is typically done by maximising the log likelihood function, which given Eq. (5a), has the form

$$\ln(L(j)) = \sum_{i=1}^{n_1(j)} \left\{ [(\lambda(j) - 0.5)\ln(\lambda(j))] - \ln\Gamma[\lambda(j)] - \ln[\sigma(j)] + \sqrt{\lambda(j)}e(ij) - \lambda e^{e(ij)/\sqrt{\lambda(j)}} \right\} + \sum_{i=1}^{n_2(j)} \ln Q\left(\lambda(j), \lambda(j)e^{e(ij)/\sqrt{\lambda(j)}}\right) \quad j=1, m \quad (5d)$$

where Q() is the incomplete gamma integral and gives the probability of specimen surviving a given length of time (natural log of time) and $n_1(j)$ are the number of failed specimens in the jth batch of data and $n_2(j)$ the number of censored or un failed specimens in the jth batch. Step 1 then involves maximising Eq.(5d) for each of the j batches of material to give the parameter estimates shown in Eqs. (3,4). This yields m separate estimates of the parameters b_0 to b_4 as well as σ (one for each batch). During this procedure L(j) may differ between the batches in which case the failure times associated with each batch are distributed differently.

4.2. Step 2

The estimated batch to batch variation in the Wilshire parameters can next be related to all the variables that define the nature of the batches and which distinguish one batch of the same material from another

$$\begin{aligned}
&\hat{b}_0(j) = c_0 + c_{0k}\sum_{k=1}^{p} x_k(j) + u_0(j),\\
&\hat{b}_1(j) = c_1 + c_{1k}\sum_{k=1}^{p} x_k(j) + u_1(j),\\
&\ldots\ldots\ldots\ldots\ldots\\
&\hat{b}_4(j) = c_4 + c_{4k}\sum_{k=1}^{p} x_k(j) + u_4(j),\\
&\hat{\tau}_1^*(j) = c_5 + c_{5k}\sum_{k=1}^{p} x_k(j) + u_5(j),\\
&\hat{\sigma}(j) = c_6 + c_{6k}\sum_{k=1}^{p} x_k(j) + u_6(j),
\end{aligned} \quad (6)$$

where $x_k(j)$ are the k =1 to p variables that describe the jth batch, e.g. $x_1(j)$ may the carbon content and $x_2(j)$ may be the chrome content of batch j. These batch specific variables can be used to predict the parameters of the Wilshire equation for each specific batch. Part of the variation in the Wilshire parameters between batches may also be random in nature, i.e. not dependent on these batch variables, and this is picked up in the error terms $u_0(j)$ to $u_6(j)$. This is likely to be the case as it is not possible to quantify all the variables that distinguish one

batch from another and these missing variables can be captured by these random error terms. Varying degrees of generality can then be achieved by specifying different properties for all these random error terms – to be discussed further below.

Eq. (5d) reveals the degrees f freedom problem involved in estimating all the parameters in Eq. (6). Typically a creep data set will be made up of no more than around m = 10 batches, so for example there will only be 10 values for say b_0. Typically, p will exceed this number of batches (for example at least 10 chemical elements are typically listed for a particular batch and then there will be further distinguishing features such as hardness, heat treatments etc.). With p exceeding the number of batches m, it becomes impossible to estimate all the c parameters in Eq. (6) as there are insufficient degrees of freedom. The solution is to use partial least squares (pls) within this second step of the estimation procedure.

In seeking dimensionality reduction useful for predictive purposes, the obvious objective criteria to use is to choose components that are linear combinations of the x's that maximise the covariance between the dependent variable and all the explanatory variables. This essentially is what partial least squares does. These pls components are typically labelled T_{1q}, T_{2q},....T_{hq} , (q = 0, 6 and h is less than m), and there are some intuitively straightforward ways to implement pls and these details are contained in Appendix A to this paper. Once the pls components are calculated, the PLS regression equations then take the form

$$\begin{aligned}
\hat{b}_0(j) &= \delta_0 + \delta_{10}T_{10}(j) + ... + \delta_{ho}T_{ho} + u_0(j),\\
\hat{b}_1(j) &= \delta_1 + \delta_{11}T_{11}(j) + ... + \delta_{h1}T_{h1} + u_1(j),\\
\hat{b}_2(j) &= \delta_2 + \delta_{12}T_{12}(j) + ... + \delta_{h2}T_{h2} + u_2(j),\\
\hat{b}_3(j) &= \delta_3 + \delta_{13}T_{13}(j) + ... + \delta_{h3}T_{h3} + u_3(j),\\
\hat{b}_4(j) &= \delta_4 + \delta_{14}T_{14}(j) + ... + \delta_{h4}T_{h4} + u_4(j),\\
\hat{\tau}_1^*(j) &= \delta_5 + \delta_{15}T_{15}(j) + ... + \delta_{h5}T_{h5} + u_5(j),\\
\hat{\sigma}(j) &= \delta_6 + \delta_{16}T_{16}(j) + ... + \delta_{h6}T_{h6} + u_6(j).
\end{aligned} \tag{7}$$

There are two ways to estimate the delta parameters in Eq. (7). The error terms u_0 to u_6 can be assumed to be normally and independently distributed in which case ordinary least squares can be applied separately to each of the equations in Eq. (7). Again the hat symbol will be used to denote these estimates

$$\hat{\delta}_0, \hat{\delta}_1, \hat{\delta}_2, \hat{\delta}_3, \hat{\delta}_4, \hat{\delta}_5, \hat{\delta}_6, \hat{\delta}_{aq} \quad ; \quad \text{q = 0, 6 and a = 1, h} \tag{8}$$

Alternatively, if the errors terms u_0 to u_6 are normally distributed with zero mean, but also correlated with each other, then in this the case values for these seven error variables are generated by a joint normal distribution with zero mean which is given by

$$\varphi[\mathbf{u}(j)] = (2\pi)^{-5/2}\left|\boldsymbol{\Sigma}\right|^{-0.5} e^{-0.5(\mathbf{u}(j))\boldsymbol{\Sigma}^{-1}(\mathbf{u}(j))'} \tag{9a}$$

In Eq. (10a) the vector **u**(j) contains the jth observation on the q = 0, 6 random error variables defined by Eq. (7) . In Eq. (9a) the matrix **Σ** is the associated covariance matrix (symmetric) containing the variances for each of the u_q errors terms down the diagonal and the covariance's between the errors off this diagonal

$$\mathbf{\Sigma} = \begin{bmatrix} \sigma^2_{u_0} & & & & \\ \sigma_{u_{01}} & \sigma^2_{u_1} & & & \\ \sigma_{u_{02}} & \sigma_{u_{12}} & \sigma^2_{u_2} & & \\ \cdots & \cdots & \cdots & \cdots & \\ \sigma_{u_{06}} & \sigma_{u_{16}} & \sigma_{u_{26}} & \sigma^2_{u_{36}} & \sigma^2_{u_6} \end{bmatrix} \qquad (9b)$$

σ^2_{u0} is the variance of the u_0(j), σ^2_{u1} is the variance of the u_1(j) and so on. σ_{u01} is the covariance between the u_0(j) and the u_1(j) values and σ_{u02} is the covariance between the u_0(j) and the u_2(j) values and so on. **Σ** is a symmetric matrix with elements above the diagonal (left blank) equalling those elements below the diagonal.

Such dependence comes under the heading of seemingly unrelated regressions (SUR) and estimation of the delta parameters in such circumstances is described in appendix B of this paper. If the errors are dependent then SUR estimation is more efficient than the separate application of ordinary least squares to each equation - i.e. it will result in smaller standard errors for the estimated delta parameters. The tilde symbol will be used to denote these SUR estimates

$$\tilde{\delta}_0, \tilde{\delta}_1, \tilde{\delta}_2, \tilde{\delta}_3, \tilde{\delta}_4, \tilde{\delta}_5, \tilde{\delta}_6, \tilde{\delta}_{aq} \qquad q = 0, 6 \quad \text{and} \quad a = 1, h \qquad (10)$$

Also, this paper will restrict its analysis to h = 1.

4.3. Percentiles

A simulation approach can also be used to obtain percentiles for this hierarchical model. This approach requires first simulating values for $\mathbf{u}_q$, using $\mathbf{u}_q = \hat{\Lambda}\mathbf{v}_q$, where $\hat{\Lambda}\Lambda^{/} = \hat{\Sigma}$ and $\mathbf{v}_q$ is a vector of seven observation chosen at random from the standard normal distribution. This yields the vector u_q containing seven randomly selected values for u_0 to u_6. The hat refers to estimated values obtained in the way described above. These simulate values for u_0 to u_6 are then inserted into Eq. (8), together with a particular batch characteristic (i.e. values for T_{hq}) to simulate values for all the b_q(j), τ^*_1(j) and σ(j) for that particular batch. All these values are then inserted into Eqs. (5b,c) together with a particular stress and temperature to obtain any desired percentile life time prediction for that batch operating at that chosen stress/temperature combination. This process is repeated many times from which it is possible to calculate the average of all these percentile predictions. Finally, repeating this process for other stress and temperatures yields a predicted relationship between time to failure and any stress/temperature combination.

5. The data

The above techniques are illustrated using 12Cr stainless steel bars for turbine blades. For m = 9 batches of this product (coded alphabetically RBA to RBJ), both the creep and creep fracture properties have been published in creep data sheet No.13b by the National Institute for Materials Science [17], Japan. Each batch has a slightly different chemical composition as shown in Table 1a and underwent different heat treatments as shown in the final column of Table 1b. Table 1b also shows some other material properties of these batches, such as Rockwell hardness and grain size, that further help characterize and describe the differences between all these batches. A total of 241 specimens were tested at constant load over a wide range of conditions: 373 MPa - 41MPa and 723K – 873K. (Recordings were also made of minimum creep rate ($\dot{e}_m$) and the times to attain various strains (t_e) at 0.005, 0.01, 0.02 and 0.05 over this range of test conditions). At the time of publication some ten of these specimens remained unfailed and so the published times are actually censored times as these tests are still ongoing. Batches RBC, REE, RBF and RBG had no such unfailed specimens.

Table 1a
Chemical composition of each batch of 12Cr stainless steel (wt%).

Table 1b
Other batch characteristics of 12Cr stainless steel.

6. Results

6.1. Ignoring batch to batch variation

Fig. 1a shows all the batches in this data set within the typical Wilshire presentation for times to failure. Also shown is the Wilshire fit to this data obtained by ignoring the batch to batch differences described in Tables 1 and which can be visualised in Fig. 1a. The censored data points are shown as solid elongated bars and these were treated as actual failure times when estimating the parameters of Eq. (2a) using ordinary least squares and a grid search for τ^*_1. In obtaining the resulting estimates shown in Fig.1a all the batches were treated as homogenous, i.e. as a single sample of data, and assuming that the log failure times are normally distributed so that the standard deviation in such log failure times is estimated at $\sigma* = 0.767$. Although not reported here, the t statistic associated with parameter b_4 in Eq. (2a) suggest that the values for u and k_1 are statistically significantly different from each other (at the 1% significance level) above and below the kink point occurring at $\tau^*_1 = -0.8125$ (or at a normalised stress of 0.642). Whilst the t statistic on parameter b_5 also suggest a significantly different activation energy either side of this kink point, the magnitude of this difference is very small (around 6 kJ mol^{-1}). This suggests that for this materials the activation energy for lattice self-diffusion is around 375 kJ mol-1. Also shown are the 0.5 and 99.5 percentiles obtained using Eqs. (5b,c) assuming that failure times are log normally distributed. This interval should contain 99% of all the data and is very wide because the batch to batch

variation is ignored or treated as homogenous. This 0.5 percentile can be used to life any batch (as it gives a replacement time such that there is only a 0.5% chance of failure before that time) and this will be compared to the results below that make use of particular batch characteristics.

Fig.1a. Plot of $\ln[-\ln(\tau/\tau_{TS})]$ against $t_F\exp[Q^*_c/RT]$ for each batch of 12Cr stainless steel.

Fig. 1b shows the Wilshire fit in Fig.1a but within the normalized stress v time to failure space for two temperatures – 773K (the lowest temperature in the data set) and 823K (the highest temperatures). Again the 0.5 and 99.5 percentiles are shown. So at 823K and a normalized stress of 0.17, there is a 50% chance of failure at or before 326 hours (the median prediction), but a 0.5% chance of failure at or before only 45 hours. Again at 723K and a normalized stress of 0.42, there is a 50% chance of failure at or before 117,329 hours (the median prediction), but a 0.5% chance of failure at or before only 16,273 hours. So if these were the operating conditions, replacement would have to be made after only 16,273 hours if the risk of failure was to be as low as 0.5%. This would be true no matter what batch was being considered. However, as shown in Fig. 1b, batch RBF has failure times consistently above the median life time prediction at both temperatures, suggesting this batch of material may be more creep resistant that the others on average. If the characteristics of this batch could be used to make prediction it would be possible to obtain more accurate median life time predictions and possibly also narrow down this 99 percentile interval and obtain safe life times that are more realistic and suitable to the batch of material being used at a specific power plant. The following sub sections show the results of trying to do just his using the methods discussed in sections 3 and 4.

Fig.1b. Plot of times to failure against the normalised stress at 873K and 723K for 12 Cr stainless steel, together with the percentile predictions obtained by ignoring batch differences.

6.2. Results of step 1 estimation

Table 2 shows the results of maximizing Eq. (5d) separately for each batch of material. The one but last column of this table shows that for most batches the log likelihood is maximized when $k = \infty$, suggesting that for most batches failure times follow a log normal distribution. The exceptions are batches RBB, RBG and RBH. However, for these batches, Fig.2 plots the difference between the log likelihood shown in Table 2 and the maximized log likelihoods for the other k values shown on the horizontal axis (actually $k^{-0.5}$ is shown on the horizontal axis to rescale the range 1 to ∞ to the narrower 0 to 1). This difference when multiplied by -2 has a chi square distribution with 1 degree of freedom under the null hypothesis that k equals the specified value shown on the horizontal axis. The 5% critical value for this chi square distribution is shown as the horizontal line in Fig.2. Only for batch RBH is this critical value exceeded, so that for batches RBB and RBG the null hypothesis of a log normal distribution cannot be rejected at the 5% significance level even though this distribution does not give the largest log likelihood. At odds with all the other batches, the log likelihood for batch RBH is maximized when k = 1 (corresponding to failure times being Weibull distributed). Further, batch RBH appears not to have a log normal failure time

distribution at the 5% significance level (but does so at the 1% significance level and so this batch to may have the same distribution as the others).

Fig. 2. Plot of k against the likelihood ratio statistic testing the null hypothesis that k takes on the values shown on the horizontal axis.

Reading down the fourth column of Table 2 shows that the activation energy varies considerably between the batches, varying from a low of 273 $kJmol^{-1}$ for RBH to a high of 331 $kJmol^{-1}$ for batch RBF. Reading down column eight of Table 2 reveals that the breakpoint in the Wilshire equation occurs at very different normalized stresses so that for batch RBB this discontinuity occurs at a normalized stress of exp(-exp(0.22)) = 0.29, whilst for batch RBJ this occurs at a normalized stress of 0.56. The average of all these varying break points is very similar to the break point estimated when ignoring batch differences, i.e. the value shown in Fig.1a. Further, column five reveals that at least the 5% significance level, the values for $b_o(j)$ and $b_1(j)$ are significantly different above and below these points of discontinuity. However, column 6 reveals that for batches RBA, RBB, RBC and RBG, the activation energy is not different above and below these points of discontinuity. For the remaining batches the activation energy does vary either side of the points of discontinuity at least the 10% significance level. But for these batches the change in the activation energy above and below the points of discontinuity are quite small – no more than 11.79 $kJmol^{-1}$.

The results down column 7 of Table 1 are informative when looking at probabilities of failure. When $k = \infty$, so that times to failure are log normally distributed, $\sigma(j)$ is the standard deviation in log times to failure for batch j. For other values of k, and so other failure time distributions, $\sigma(j)$ is a multiple of the standard deviation in log failure times. What column 7 reveals therefore is that the spread or scatter present in times to failure varies quite considerably between al the batches with batch RBH having the smallest scatter in the measured failure times and batch RBD the largest. This suggests that the predictions shown in Fig.1b from the Wilshire equation that ignores the differences between batches are meaningless because these differing $\sigma(j)$ values suggest the time such that there is only a 0.5% change of failure will be much further below the median predictions for batch RBD compared to batch RBH. It makes little sense to have a predicted time corresponding to a 0.5% probability of failure that is the same for all batches – and this is also true for any percentile failure prediction. Users of 12Cr stainless steel can obtain more sensible failure time predictions based on the characteristics of the batches of material that they use.

The effect of ignoring censored data points or treating them as actual failure times can be seen by looking at, for example, Batch RBA which has three censored observations (which makes up 12% of the sample observations on this batch). Table 1 gives the estimates when these observations are treated correctly by amending the log likelihood to include the cumulative density function for such data points. If the censored data points are treated as actual failure times the maximum likelihood estimates of the parameters of the Wilshire equation change to:

$$\ln[t_f(ij)] = -26.464 + 9.794\tau^*(ij) + 291.345\left[\frac{1}{T(ij)}\right] - 2.524[\tau^*(ij) - \tau_1^*(j)]D(ij) - 1.822\left[\frac{D(ij)}{T(ij)}\right]; \quad j = 1$$

with $\sigma(1) = 0.290$.

Thus incorrectly dealing with censored data leads to an underestimation of $\sigma(1)$ and thus any percentile bands for times to failure. It also leads to an overestimation of the activation energy (inflating it by around 6 $kJmol^{-1}$), and also the values for b_o and b_1.

Table 2
Maximum likelihood estimates of Wilshire parameters: batch by batch.

6.*3. Results of step 2 estimation*

In this section equations are estimated to relate the b values in Table 2 to the batch characteristics summarized in Tables 1. Table 3 shows the results of regressing the estimated Wilshire parameters for each batch (as shown in Table 2) on the batch variables shown in Tables 1 – one at a time. The differences in these parameters between the batches is one way of visualizing the effect of batch characteristics on creep properties such as the time to failure. This was done separately for each batch variable to give the loading values shown in Table 3 – which are the $w_{1k}d_{1kq}$ values defined by Eq. (A2). Thus each loading is the best fit slope of a line within a plot of $b_q(j)$ on each x_k. The coefficient of determination, or R^2 value, of such a fit reveals how important that batch variable is in explaining the Wilshire parameters. So in Fig.3, this R^2 value is square rooted to reveal the correlation coefficient and then standardized by diving it by the standard deviation for a correlation coefficient to give a t value

$$t = [r(n-2)/1-r^2]^{0.5} \qquad (11)$$

where r is the square root of the coefficient of determination. Any standardized correlation coefficient in excess of approximately 2 is then significant at the 5% significance level so revealing the important batch variables. This t value is shown on the vertical axis of the graphs in Fig,3 – one such t value for each batch variable shown on the horizontal axis.

Fig. 3 shows that the value for b_0 and the activation energy (b_2) is predominantly determined by x_7 and x_{17} which are the P content of the batch and whether that batch went through heat treatment 3. The results indicate that an increase in P will shorten the time to failure through its influence on b_0 and the activation energy. Phosphorous is an impurity that has been shown by Takamatsu et. al. [21] to reduce creep strength in Grade 92 steel. These authors have shown that this effect occurs because Phosphorous decreases the interfacial energy at the grain boundary.

Unsurprisingly, b_3 is determined by the same variables as this parameter measure the change in b_1 either side of the point of discontinuity. The value for b_1 is predominantly determined by $x_{2,}$ x_6 and x_{15} which are the Rockwell hardness, Mn content of the batch and whether that batch went through heat treatment 1. The results indicate that an increase in Mn

will lengthen the time to failure through its influence on b_1. The effect of Mn on creep life is similar to that for Carbon, in that it plays a role in the elimination of thermally unstable M23 C6 carbides thereby increasing the time taken to reach failure. This, for example, was demonstrated by Taneike et. al. [22] in heat resistant steels for power plants.

The breakpoint does not seem to be determined by any of the batch characteristics so the variation in this breakpoint between the batch must be just random in nature. σ or the variation in failure times for each batch, is predominantly determined by x_2, x_7 x_{12} and x_{16} which are the Rockwell hardness of the batch, the P content of the batch, the Cu content of the batch and whether that batch went through heat treatment 2.

Fig. 3. Plot of the standardised correlation coefficients (given by Eq. (11)) between each Wilshire parameter and each batch variable.

These loading were then used to form the q partial least squares components T_{1q} given by Eq. (A2). Fig. 4 plots these components against all the Wilshire parameters and it can be seen that these components are very strong predictors of the batch to batch variations in these Wilshire parameters. The best fit lines shown in Fig. 4 were obtained by applying least squares separately to each Wilshire parameter and the standard errors associated with the intercept and slope of these best fit lines are shown in round brackets.. Thus T_{10} can explain 85.6% of the batch to batch variation in b_0, so the remaining variation is random in nature and quantified by the variance of the residuals in this plot, i.e. $\sigma^2_{uo} = 2.34$. b_1 has a slightly weaker systematic component with 72.7% of the batch to batch variation in b_1 being explained by T_{11}, with the remaining random variation quantified by $\sigma^2_{u1} = 0.512$. The activation energy (b_2) has a very strong systematic component with 84.6% of the batch to batch variation in b_2 being explained by T_{12} with the remaining random variation measured by $\sigma^2_{u2} = 109.5$. The scale parameter σ also has a very strong systematic component with 83.0% of the batch to batch variation in s being explained by T_{16} with the remaining random variation measured by $\sigma^2_{u6} = 0.01$. Even the break point seems to be well predicted by the partial least squares component T_{15}.

Table 3
The loadings in partial least squares components for each Wilshire parameter.

Fig. 4. Plot of the parameters of the Wilshire equation for each batch against their partial least squares components.

The SUR estimates of the best fit lines through the data points shown in Fig. 4 are shown in Eq. (12a). In comparison to the least squares estimates in Fig.4, it is apparent that the intercepts are unchanged, but the slope estimates are slightly different. The slopes are slightly lower for b_0 and b_1, but slightly higher for b_2, b_3 and b_4, but almost identical for the breakpoint and σ. The main difference comes however in the standard deviation from the random component terms u_0 to u_6. The estimated variance covariance matrix for these error terms is shown in Eq. (12b). In Eq. (12b) the values above the diagonal, shown in bold, are

actually the correlation coefficients between the various random error terms. It is clear that some of these correlations are quite large. For example, the correlation coefficient between the random components u_o and u_2 is -0.98, whilst the correlation coefficient between u_2 and u_6 is 0.71. As a consequence of this it is unsurprising to see the substantial reduction in the standard errors of the parameters that comes from account for this cross equation correlation using SUR estimation. For example, the SUR estimate for the standard error of the slope coefficient in front of component T_{12} is 0.123 (see Eq. (12a)) compared to the least squares estimate of 3.49 shown in Fig. 4. This improved efficiency holds for all the slope parameters. Thus the SUR estimates are more efficient and the parameters estimated more reliably and so these will be used in the next sub section to derive the failure time predictions for specific batches.

$$\hat{b}_0(j) = -27.596 + 4.958T_{10}(j) + u_0(j)$$
$$(0.450) \quad (0.099),$$
$$\hat{b}_1(j) = 7.76 + 3.755T_{11}(j) + u_1(j)$$
$$(0.21) \quad (0.252),$$
$$\hat{b}_2(j) = 293.992 + 4.935T_{12}(j) + u_2(j)$$
$$(3.076) \quad (0.123),$$
$$\hat{b}_3(j) = -0.883 + 3.916T_{13}(j) + u_3(j)$$
$$(0.381) \quad (0.174),$$
$$\hat{b}_4(j) = -1.825 + 4.029T_{14}(j) + u_4(j)$$
$$(0.715) \quad (0.249),$$
$$\hat{\tau}_1^*(j) = -0.251 + 4.164T_{15}(j) + u_5(j)$$
$$(0.046) \quad (0.158),$$
$$\hat{\sigma}(j) = 0.376 + 4.007T_{16}(j) + u_6(j)$$
$$(0.030) \quad (0.076). \qquad (12a)$$

Notice also that the estimated variance of the random error terms u_0 to u_6 are also lower using the SUR estimation procedure. For example, using ordinary least squares, the variance of u_0 is estimated at 2.34, but using SUR it is estimated as 1.82. As another illustration, , the variance of u_2 is estimated at 109.476, but using SUR it is estimated as 85.239. This reduction will have big implication for the estimated percentile time to failure predictions, with least squares estimates resulting in an overestimate of the range of failure times making up say the percentile interval 0.5% to 99.5%

$$\boldsymbol{\Sigma} = \begin{bmatrix} \sigma^2_{u_0} & & & & \\ \sigma_{u_{01}} & \sigma^2_{u_1} & & & \\ \sigma_{u_{02}} & \sigma_{u_{12}} & \sigma^2_{u_2} & & \\ \cdots & \cdots & \cdots & \cdots & \\ \sigma_{u_{06}} & \sigma_{u_{16}} & \sigma_{u_{26}} & \sigma^2_{u_{36}} & \sigma^2_{u_6} \end{bmatrix} = \begin{bmatrix} 1.820 & \mathbf{0.46} & \mathbf{-0.89} & \mathbf{0.23} & \mathbf{-0.06} & \mathbf{-0.31} & \mathbf{-0.38} \\ 0.395 & 0.402 & \mathbf{-0.04} & \mathbf{-0.40} & \mathbf{-0.64} & \mathbf{-0.38} & \mathbf{0.39} \\ -11.113 & -0.249 & 85.239 & \mathbf{-0.45} & \mathbf{-0.31} & \mathbf{0.06} & \mathbf{0.71} \\ 0.353 & -0.289 & -4.781 & 1.307 & \mathbf{0.11} & \mathbf{-0.41} & \mathbf{-0.55} \\ -0.179 & -0.868 & -6.228 & 0.276 & 4.639 & \mathbf{0.77} & \mathbf{-0.70} \\ -0.057 & -0.033 & 0.080 & -0.064 & 0.229 & 0.019 & \mathbf{-0.41} \\ -0.046 & 0.022 & 0.589 & -0.056 & -0.134 & -0.005 & 0.008 \end{bmatrix} \quad (12b)$$

6.4. Comparison of prediction intervals

Figs. 5 show the application of the above technique to batches RBF (j = 6) and RBH (j = 8) using the SUR and maximum likelihood estimates shown in the previous two sub sections. The solid curves in both these figures correspond to those shown in Fig. 1b and so correspond to predictions obtained using the Wilshire equation ignoring batch specific characteristics. In Fig. 5a the failure times for batch RBH are shown for two separate temperatures. Notice that at temperature 723K the median failure time prediction obtained when ignoring the batch specifics of RBH underestimates the actual failure times for specimens cut from this batch. However, when the batch characteristics of RBH are combined with the loading values in Table 3, specific values for T_{11} to T_{16} are obtained, which yield predictions of the Wilshire parameters for that batch:

$$\begin{aligned}
\hat{b}_0(6) &= \text{-}27.596 + 4.958(0.6709) + u_0(6) \\
&\quad (0.450) \quad (0.099), \\
\hat{b}_1(6) &= 7.76 + 3.755(0.073) + u_1(6) \\
&\quad (0.21) \ (0.252), \\
\hat{b}_2(6) &= 293.992 + 4.935(-4.3922) + u_2(6) \\
&\quad (3.076) \quad (0.123), \\
\hat{b}_3(6) &= \text{-}0.883 + 3.916(0.039) + u_3(6) \\
&\quad (0.381) \ (0.174), \\
\hat{b}_4(6) &= \text{-}1.825 + 4.029(1.0154) + u_4(6) \\
&\quad (0.715) \ (0.249), \\
\hat{\tau}_1^*(6) &= \text{-}0.251 + 4.164(\text{-}0.0305) + u_5(6) \\
&\quad (0.046) \ (0.158), \\
\hat{\sigma}(6) &= 0.376 + 4.007(\text{-}0.0581) + u_6(6) \\
&\quad (0.030) \ (0.076).
\end{aligned} \quad (13)$$

where for example T_{11} = 0.6709 = 1.2580*(4.8-5.98)+……..-0.6254*(0-0.11) and where 5.98 is the average C content over all nine batches, 4.8 is the C content for batch RBH, 1.2580 the loading in front of C shown in the top row of the second column of Table 3, 0 is the dummy variable for heat treatment 5 for batch RBH and 0.11 the average value for this dummy variable over the nine batches.

Using the procedure described in sub section 4.4, random values are obtained for $u_0(6)$ to $u_6(6)$ which are substituted into Eq. (13) to yield values for $b_0(6)$ to $\sigma(6)$. These are then inserted into Eq. (5b), to obtain a median, 0.5 and 99.5 percentile failure time predictions at a specified normalised stress and temperature. Then this process of simulating values for $u_0(6)$ to $u_6(6)$ is repeated 2,000 times to obtain 2000 median, 0.5 and 99.5 percentile failure time predictions. These are then averaged out to obtain the median, 0.5 and 99.5 percentile failure time predictions. The dashed curves in Fig. 5a show these median and percentile predictions when this process is repeated over a range of normalised stresses and two temperatures.

Fig.5a. Plot of times to failure against the normalised stress at 873K and 723K for batch RBH of 12Cr stainless steel, together with the percentile predictions obtained by a. ignoring batch differences (solid curves) and b. accounting for batch differences (dashed curves).

It can be seen that by taking into account the batch characteristics of RBH, the median prediction is now much more representative of this batches actual failure times than is the median prediction obtained by ignoring these characteristics. Notice also that the 0.5 percentile prediction is also associated with much higher failure times than that obtained by ignoring the batch characteristics of RBH. This suggest that for companies operating with a 0.5% risk of failure, the materials could stay in operation much longer than that suggested from the use of the Wilshire equation that ignores batch specific characteristics. This in turn would have significant financial benefits. The 99.5 – 0.5 percentile prediction band is much narrower when considering the batch characteristics for RBH compared to ignoring batch variation, because as shown in Table 2 $\sigma(6) = 0.1449$, which is substantially lower than $\sigma^* = 0.767$ shown in Fig. 1a where batch to batch variation is ignored (which can be considered an weighted average of all the σ values shown in Table 2). This band is also much narrower at 873K as well and interestingly the point of discontinuity occurs at a much lower normalised stress in comparison to when batch characteristics are ignored.

Fig. 5b repeats this analysis for batch RBF. Again notice that taking into account the characteristics of this batch produces much more realistic median time predictions than simply ignoring such characteristics. The 99.5 – 0.5 percentile prediction band is again a little narrower than that obtained when ignoring batch to batch variation. However this band is not as narrow as for batch RBH because $\sigma(8) = 0.5339$ which is higher than $\sigma(6)$ but now much closer to $\sigma^* = 0.767$ shown in Fig. 1a - where batch to batch variation is ignored. The point of discontinuity occurs at a slightly lower normalised stress and breaks in the opposite direction in comparison to when batch characteristics are ignored.

It is clear that to have predictions of safe life (e.g. a time such that there is a small chance of failure before then – such as 0.5%) that are suitable for a batch of material actually being used at say a power planet, the batch to batch variation cannot be ignored. It is more realistic to take into account the characteristics of that batch when making such predictions.

Fig.5b. Plot of times to failure against the normalised stress at 873K and 723K for batch RBF of 12Cr stainless steel, together with the percentile predictions obtained by a. ignoring batch differences (solid curves) and b. accounting for batch differences (dashed curves).

7. Conclusions

Specific batch characteristics were incorporated into the Wilshire equation for times to failure using a new twostep estimation procedure that made use of maximum likelihood at Seemingly unrelated regression (SUR) techniques. These specific batch characteristics included the batches chemical composition, the heat treatment it had been subjected to, the grain size, the frequency of inclusions and its hardness. Applying this modified Wilshire equation to 12Cr stainless steel lead to the following results:

(1) For all but one batch of material, the null hypothesis that failure times were log normally distributed could not be rejected at the 5% significance level. However, for batch RBH the log likelihood function was maximised when failure times were Weibull distributed (although at the 1% significance level the null hypothesis that failure times were log normally distributed could not be rejected).

(2) There was considerable variation in the values for the Wilshire parameters between all the batches, with the point of discontinuity also varying from batch to batch. For example, the estimated activation energy varied between the limits of 273 $kJmol^{-1}$ for RBH to a high of 331 $kJmol^{-1}$ for batch RBF.

(3) The batches had very different standard deviations in the log failure times with σ being largest for batch RBD and smallest for batch RBH

(4) The variation in the Wilshire parameters between the batches had both a systematic and a random component. For the systematic component only a few batch characteristics seemed to be important in explaining this variation – namely the P, Mn and Cu content of the batch, the batches hardness and some types of heat treatment. The random component was modelled as a multivariate normal distribution with the SUR estimates producing more efficient parameters estimates and smaller estimates for the size of the random component.

(5) Result 3 meant that failure time prediction obtained from the Wilshire equation that ignored individual batch characteristics were irrelevant for a specific batch of material. Using the Wilshire equation modified to account for batch characteristics produced median predictions that were more representative of particular batches of data and the 0.5 - 99.5 percentile bands were much narrower and so the lower bound provided a more economically feasible safe life. The modification Wilshire equation should therefore allow for the more reliable safe life determination of specific batches actually being used by, for example, power generating companies.

Conflict of interest

There are no relationships or interests of this manuscripts author that could influence or bias the submitted work.

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Appendix

A. Partial least squares (pls)

In the second step of the estimation procedure there are q = 0, 6 dependent variables – $b_0(j)$ to $b_4(j)$, $\tau^*_1(j)$ and $\sigma(j)$ with j = 1 to m observations on each. If $b_5(j) = \tau^*_1(j)$ and $b_6(j) = \sigma(j)$ then there is a sample of size m from which to estimate a linear relationship between each b_q and the explanatory variables $x_1, x_2, \ldots, x_p$ which distinguish the m batches from one another. For j = 1, ..., m, and q = 0,…., 6, the jth datum in the sample is denoted by $\{x_1(j), \ldots, x_p(j), b_q(j)\}$. Denote their sample means by $\bar{b}_q = \Sigma_j b_q(j)/m$ and $\bar{x}_k = \Sigma_j x_k(j)/m$, for k = 1,….,p. To simplify notation, each b_q and all the x_k are next centred to give variables $u_{1q}(j)$ and $v_{1k}(j)$, where $u_{1q}(j) = b_q(j) - \bar{b}_q$ and, $v_{1k}(j) = x_k(j) - \bar{x}_k$. The sample means of u_{1q} and u_{1k} are therefore by construction zero.

The partial least squares components can now be determined sequentially [23]. The first and most important components, T_{1q}, are intended to be useful for predicting the u_{1q} and are constructed as a linear combination of the v_{1k}'s. During its construction, sample correlations between the v_{1k}'s are ignored. To obtain T_{1q} for each q, u_{1q} is first regressed against v_{11}, then against v_{12}, and so on for each v_{1k} in turn. Sample means are zero, so for q = 0,…,6, and k = 1, . .. , p the resulting least squares regression equations are

$$u_{1q}(j) = d_{1kq} v_{1k}(j) + \eta_{1q}(j) \qquad \text{with} \qquad d_{1kq} = \frac{\sum_{j=1}^{m} u_{1q}(j) v_{1k}(j)}{\sum_{j=1}^{m} v_{1k}^2(j)} \tag{A1}$$

where $\eta_{1q}(j)$ are a random error terms. Given q and values of the $v_{1k}(j)$, each of the p equations in Eq. (A1) provides a prediction for $u_{lq}(j)$. To reconcile these predictions, while ignoring interrelationships between the $v_{1k}(j)$, a simple average, $\Sigma_k d_{1k} v_{1k}(j)/p$ or, more generally, a weighted average can be used

$$T_{1q}(j) = \sum_{k=1}^{p} w_{1k} d_{1kq} v_{1k}(j) \qquad \text{(A2)}$$

where w_{1k} are the weights, In the true spirit of pls these weights will be inversely proportional to the variances of the d_{1kq}'s , namely $w_{1k} = (m-1)\text{var}(v_{1k})$, where $\text{var}(v_{1k})$ stands for variance of v_{1k}. An obvious alternative weighting policy is to set each w_{1k} equal to 1/p, so that each predictor of $u_{1q}(j)$ is given equal weight. This seems a natural choice and is also in the spirit of pls, which aims to spread the load among the x_k variables in making predictions. This equal weighting policy is the one adopted in this paper.

The procedure extends iteratively in a natural way to give components T_{2q}, . . ., T_{hq}, where h < m to preserve degrees of freedom and where each component is determined from the residuals of regressions on the preceding component, with residual variability in b_q being related to residual information in the x's. Specifically, suppose that $T_a(j)$ (a = 1) has just been constructed from variables $u_{lq}(j)$ and $v_{1k}(j)$, (k = 1., ..., p). To obtain $T_{(a+1)q}(j)$, first the $v_{(a+l)k}(j)$'s and $u_{a+1q}(j)$ are determined. For k = 1, ... p, $v_{ak}(j)$ is regressed against $T_{aq}(j)$, giving

$$\sum_{j=1}^{m} T_{aq}(j) v_{ak}(j) \Big/ \sum_{j=1}^{m} v_{ak}^2(j)$$

as the regression coefficient, and $V_{(a+l)k}(j)$ is defined by

$$v_{(a+1)k}(j) = v_{ak}(j) - \left\{ \sum_{j=1}^{m} T_{aq}(j) v_{ak}(j) \Big/ \sum_{j=1}^{m} v_{ak}^2(j) \right\} T_{aq}(j) \qquad \text{(A3)}$$

Its sample values, $v_{(a+1)k}(j)$, are the residuals from the regression. Similarly, $u_{(a+1)q}(j)$ is defined by

$$u_{(a+1)q} = u_{aq}(j) - \left\{ \sum_{j=1}^{m} T_{aq}(j) u_{aq}(j) \Big/ \sum_{j=1}^{m} T_{aq}^2(j) \right\} T_{aq}(j)$$

where $u_{(a+1)q}(j)$, are the residuals from the regression of $u_{aq}(j)$ on $T_{aq}(j)$.

The "residual variability" in b_q is $u_{(a+1)q}(j)$ and the "residual information" in x_k is $v_{(a+l)k}(j)$, so the next stage is to regress $u_{(a+1)q}(j)$ against each $v_{(a+l)k}(j)$ in turn. The pth regression yields $d_{(a+1)kq} v_{(a+1)k}$ as a predictor of $u_{a+1q}(j)$, where

$$d_{(a+1)kq} = \sum_{j=1}^{m} u_{(a+1)q}(j) v_{(a+1)k}(j) \Big/ \sum_{j=1}^{m} v_{(a+1)k}^2(j) \qquad \text{(A4)}$$

Forming a linear combination of these predictors, as in Eq. (A2), gives the next component

$$T_{(a+1)q}(j) = \sum_{k=1}^{p} w_{(a+1)k} d_{(a+1)kq} v_{(a+1)k}(j) \tag{A5}$$

The PLS regression equation is then of the form

$$b_q(j) = \delta_0 + \delta_1 T_{1q}(j) + \delta_2 T_{2q}(j) + ... + \delta_h T_{hq}(j) + u_q(j) \tag{A6}$$

where each of the components $T_{aq}(j)$ are a linear combination of all the x_k.

B. SUR estimation

SUR estimation is a feasible generalised least squares procedure that is iterated to convergence [24]. First, ordinary least squares is applied separately to the 7 equations in Eq. (7). This yields the least squares estimates for all the delta parameters (denoted with a hat above each delta) from which least squares estimates of the u_q variables can be obtained – again denoted with a hat

$$\begin{aligned}
&\hat{b}_0(j) - [\hat{\delta}_0 + \hat{\delta}_{10} T_{10}(j) + ... + \hat{\delta}_{ho} T_{ho}] = \hat{u}_0(j),\\
&\hat{b}_1(j) - [\hat{\delta}_1 + \hat{\delta}_{11} T_{11}(j) + ... + \hat{\delta}_{h1} T_{h1}] = \hat{u}_1(j),\\
&\hat{b}_2(j) - [\hat{\delta}_2 + \hat{\delta}_{12} T_{12}(j) + ... + \hat{\delta}_{h2} T_{h2}] = \hat{u}_2(j),\\
&\hat{b}_3(j) - [\hat{\delta}_3 + \hat{\delta}_{13} T_{13}(j) + ... + \hat{\delta}_{h3} T_{h3}] = \hat{u}_3(j),\\
&\hat{b}_4(j) - [\hat{\delta}_4 + \hat{\delta}_{14} T_{14}(j) + ... + \hat{\delta}_{h4} T_{h4}] = \hat{u}_4(j),\\
&\hat{\tau}_1^*(j) - [\hat{\delta}_5 + \hat{\delta}_{15} T_{15}(j) + ... + \hat{\delta}_{h5} T_{h5}] = \hat{u}_5(j),\\
&\hat{\sigma}(j) - [\hat{\delta}_6 + \hat{\delta}_{16} T_{16}(j) + ... + \hat{\delta}_{h6} T_{h6}] = \hat{u}_6(j).
\end{aligned} \tag{B1}$$

Next, these estimated values for u_q can be used to estimate the elements of $\boldsymbol{\Sigma}$ in Eq. (9b) using the standard formulas for the sample variance and covariance

$$\hat{\sigma}_{uq}^2 = \frac{\sum_{j=1}^{m} u_q^2(j)}{m-1} \text{ and } \hat{\sigma}_{uqv} = \frac{\sum_{j=1}^{m} u_q(j) u_v(j)}{m-1} \quad q = 0, 6 \text{ and } v = q + 1 \tag{B2}$$

given that u_q are zero mean variables. Eq. (B2) can be used to estimate $\boldsymbol{\Sigma}$

$$\hat{\boldsymbol{\Sigma}} = \begin{bmatrix}
\hat{\sigma}_{u_0}^2 & & & & \\
\hat{\sigma}_{u_{01}} & \hat{\sigma}_{u_1}^2 & & & \\
\hat{\sigma}_{u_{02}} & \hat{\sigma}_{u_{12}} & \hat{\sigma}_{u_2}^2 & & \\
... & ... & ... & ... & \\
\hat{\sigma}_{u_{06}} & \hat{\sigma}_{u_{16}} & \hat{\sigma}_{u_{26}} & \hat{\sigma}_{u_{36}}^2 & \hat{\sigma}_{u_6}^2
\end{bmatrix} \tag{B3}$$

The generalised regression model then applies to the stacked model

$$\begin{bmatrix}\hat{\mathbf{b}}_0\\ \hat{\mathbf{b}}_1\\ :\\ \hat{\mathbf{b}}_6\end{bmatrix}=\begin{bmatrix}\mathbf{T}_0 & \mathbf{0} & .. & \mathbf{0}\\ \mathbf{0} & \mathbf{T}_1 & .. & \mathbf{0}\\ : & & : & \\ \mathbf{0} & \mathbf{0} & .. & \mathbf{T}_6\end{bmatrix}\begin{bmatrix}\boldsymbol{\delta}_0\\ \boldsymbol{\delta}_1\\ :\\ \boldsymbol{\delta}_6\end{bmatrix}+\begin{bmatrix}\mathbf{u}_0\\ \mathbf{u}_1\\ :\\ \mathbf{u}_6\end{bmatrix} \quad ; \quad \text{or} \quad \mathbf{b}=\mathbf{T}\boldsymbol{\delta}+\mathbf{u} \qquad \text{(B4)}$$

Each term in Eq. (B4) is itself a matrix hence the name stacked model. So, $\boldsymbol{\delta}_q$ is a 7x1 vector containing all the delta parameters in the q+1th row of Eq. (7). For example, $\boldsymbol{\delta}_0=\begin{bmatrix}\delta_0 & \delta_{10} & .... & \delta_{h0}\end{bmatrix}'$. $\hat{\mathbf{b}}_q$ is a m x 1 vector containing the j = 1, m values for the first step estimated b parameters shown in the q+1th row of Eq. (6) – left hand side. For example, $\hat{\mathbf{b}}_0=\begin{bmatrix}\hat{b}_0(1) & \hat{b}_0(2) & .... & \hat{b}_0(m)\end{bmatrix}'$. $\mathbf{T}_q$ is a m x (h+1) matrix containing the j = 1, m values for all the a=1, h partial least squares components that are shown in the q+1th row of Eq. (7) – right hand side. For example,

$$\mathbf{T}_0=\begin{bmatrix}1 & T_{10}(1) & ... & T_{h0}(1)\\ 1 & T_{10}(2) & ... & T_{h0}(2)\\ ... & ... & ... & ...\\ 1 & T_{10}(m) & ... & T_{h0}(m)\end{bmatrix} \quad ; \quad \mathbf{T}_1=\begin{bmatrix}1 & T_{11}(1) & ... & T_{h1}(1)\\ 1 & T_{11}(2) & ... & T_{h1}(2)\\ ... & ... & ... & ...\\ 1 & T_{11}(m) & ... & T_{h1}(m)\end{bmatrix} \text{ etc.}$$

$\hat{\Sigma}$ can now be used to form the SUR estimate for the delta parameters in Eq. (7)

$$\boldsymbol{\delta}=\left(\mathbf{T}'\hat{\Sigma}\mathbf{T}\right)^{-1}\mathbf{T}'\hat{\Sigma}^{-1}\hat{\mathbf{b}} \qquad \text{(B5)}$$

These delta estimates given by Eq. (B5) can then be used to re-estimate the u_q and thus $\hat{\Sigma}$ in Eq. (B3). In turn this can be inserted into Eq. (B5) to provide a further update of the delta parameters. This iterative process is then be repeated until convergence.

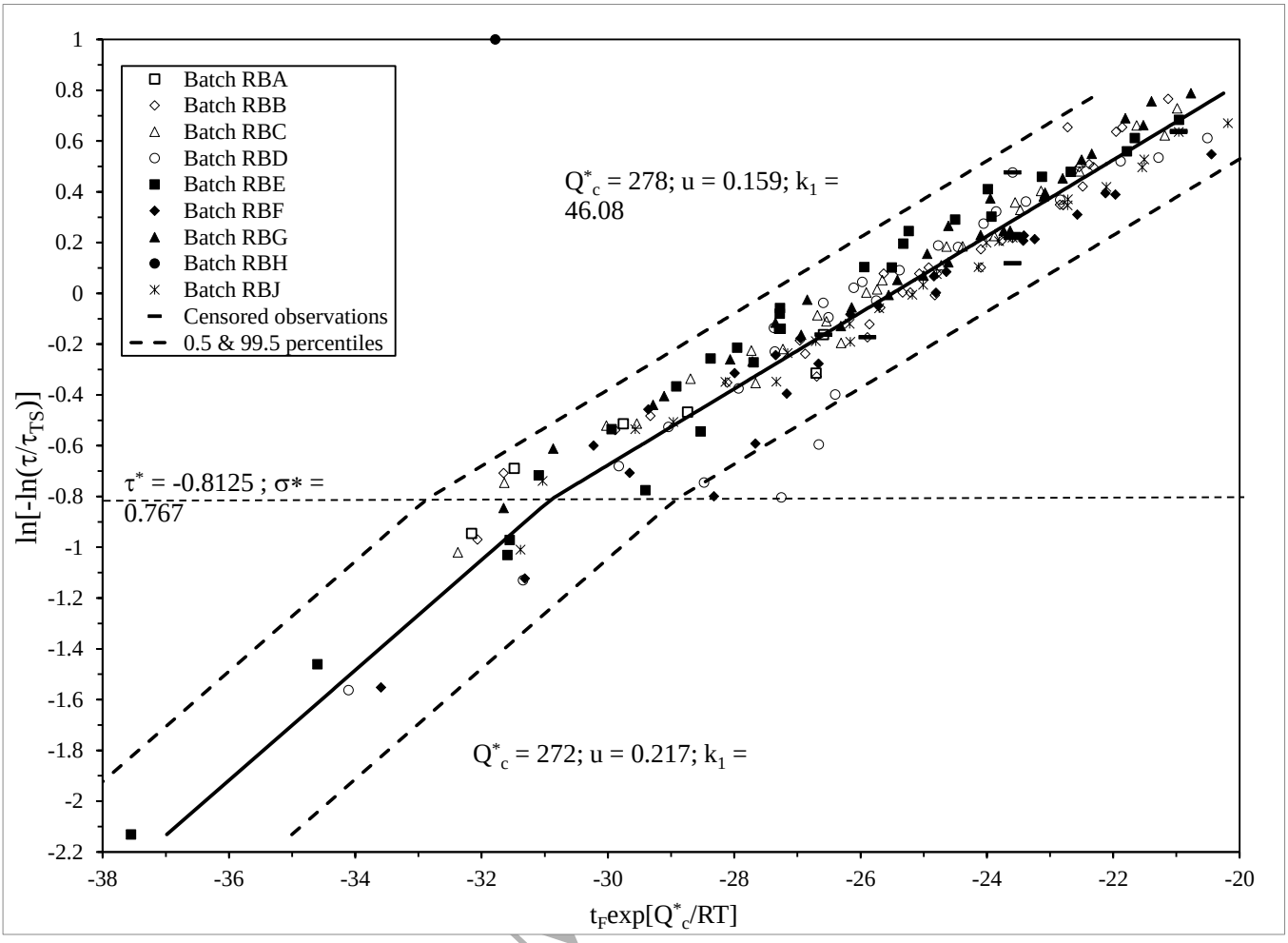


Fig.1a. Plot of $\ln[-\ln(\tau/\tau_{TS})]$ against $t_F\exp[Q^*_c/RT]$ for each batch of 12Cr stainless steel.

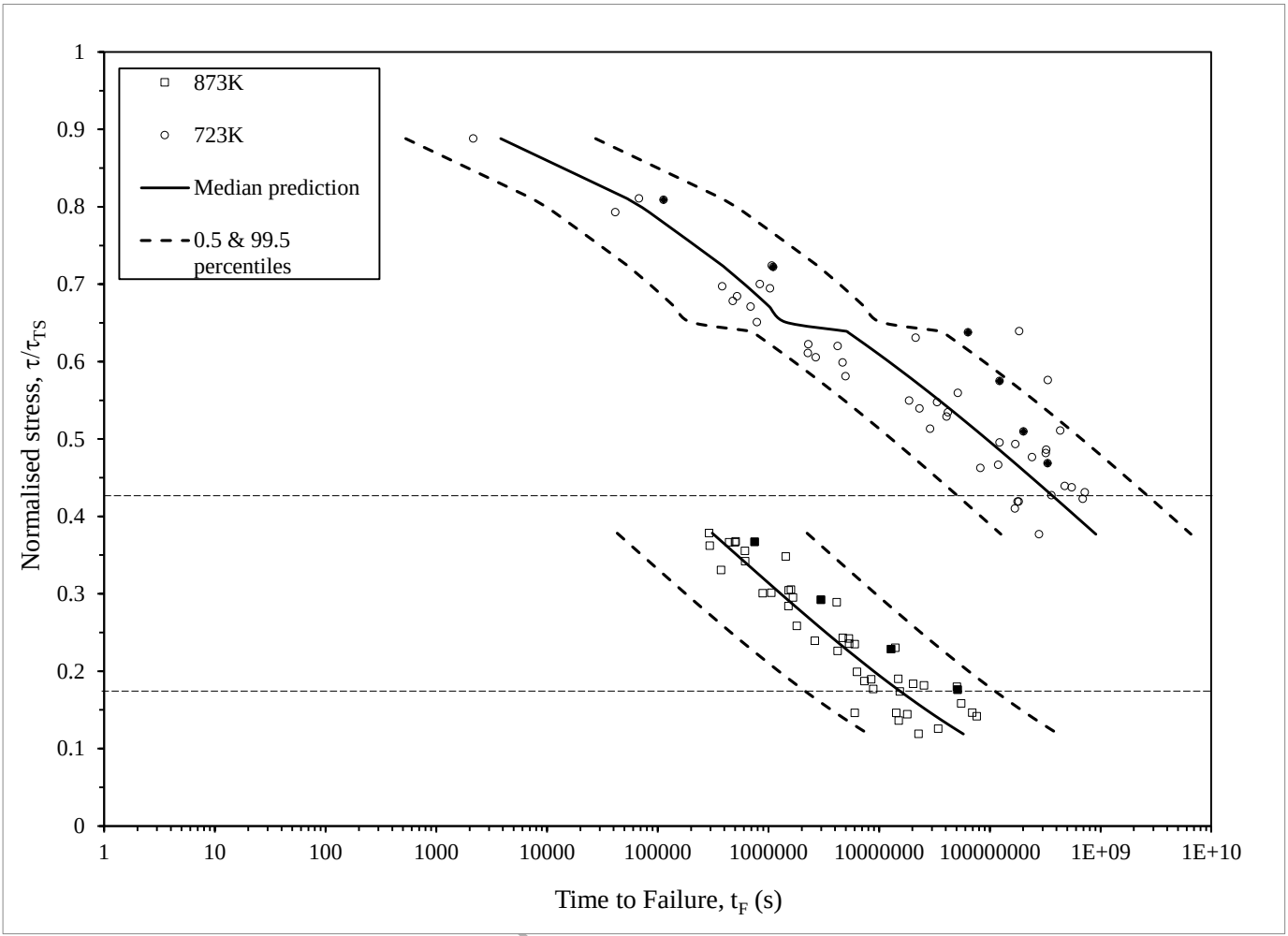


Fig.1b. Plot of times to failure against the normalised stress at 873K and 723K for 12 Cr stainless steel, together with the percentile predictions obtained by ignoring batch differences.

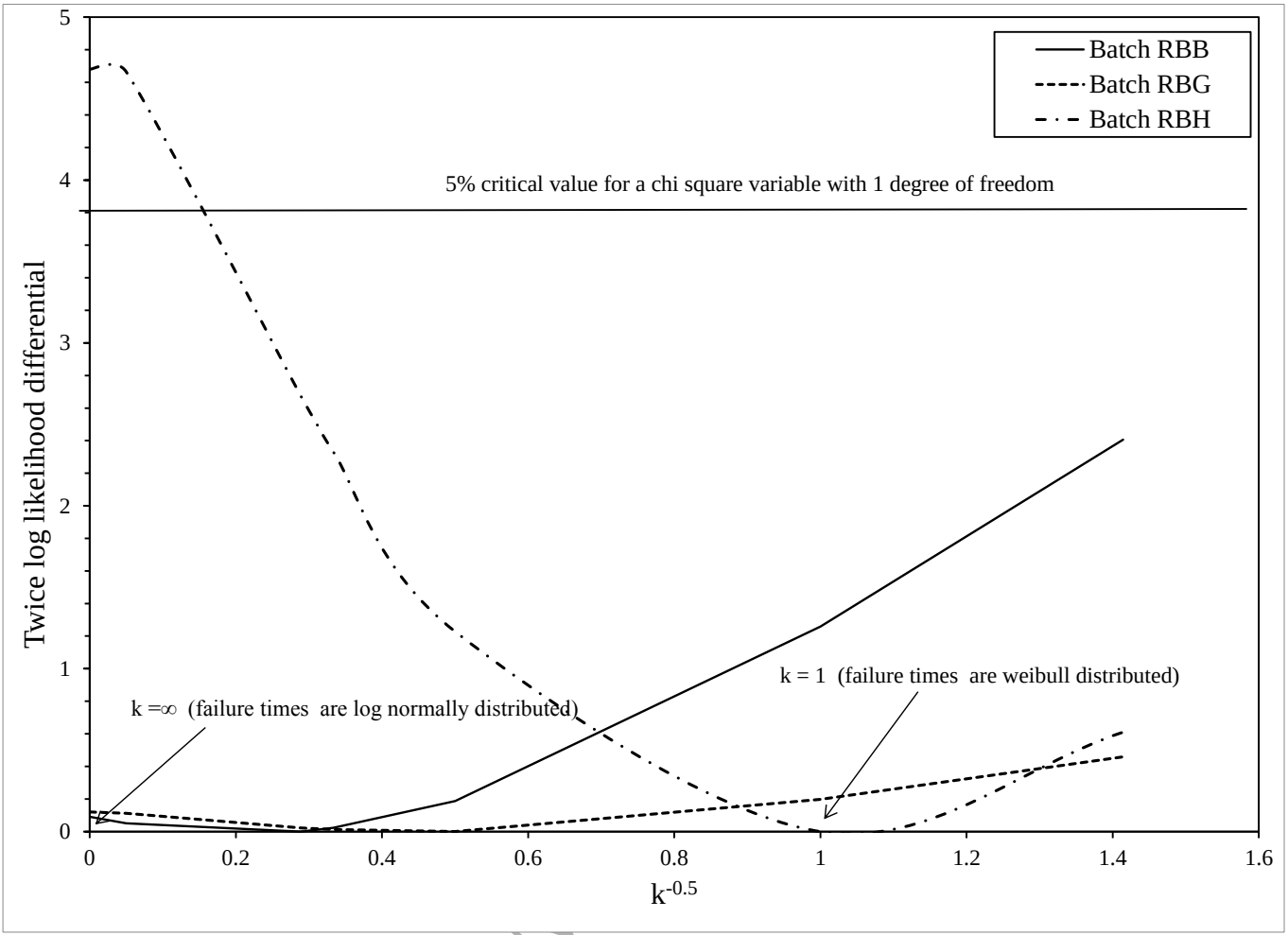
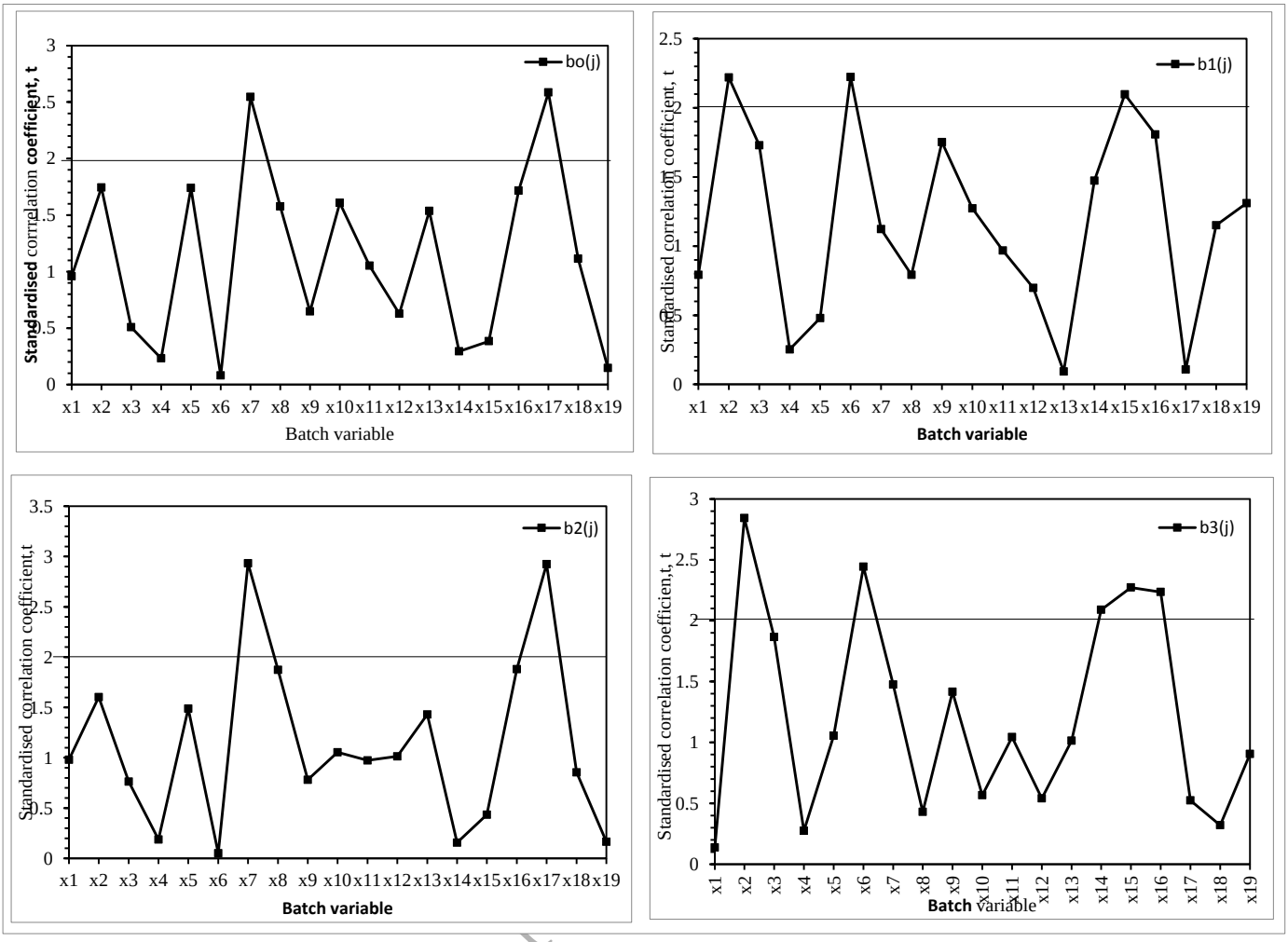


Fig. 2. Plot of k against the likelihood ratio statistic testing the null hypothesis that k takes on the values shown on the horizontal axis.



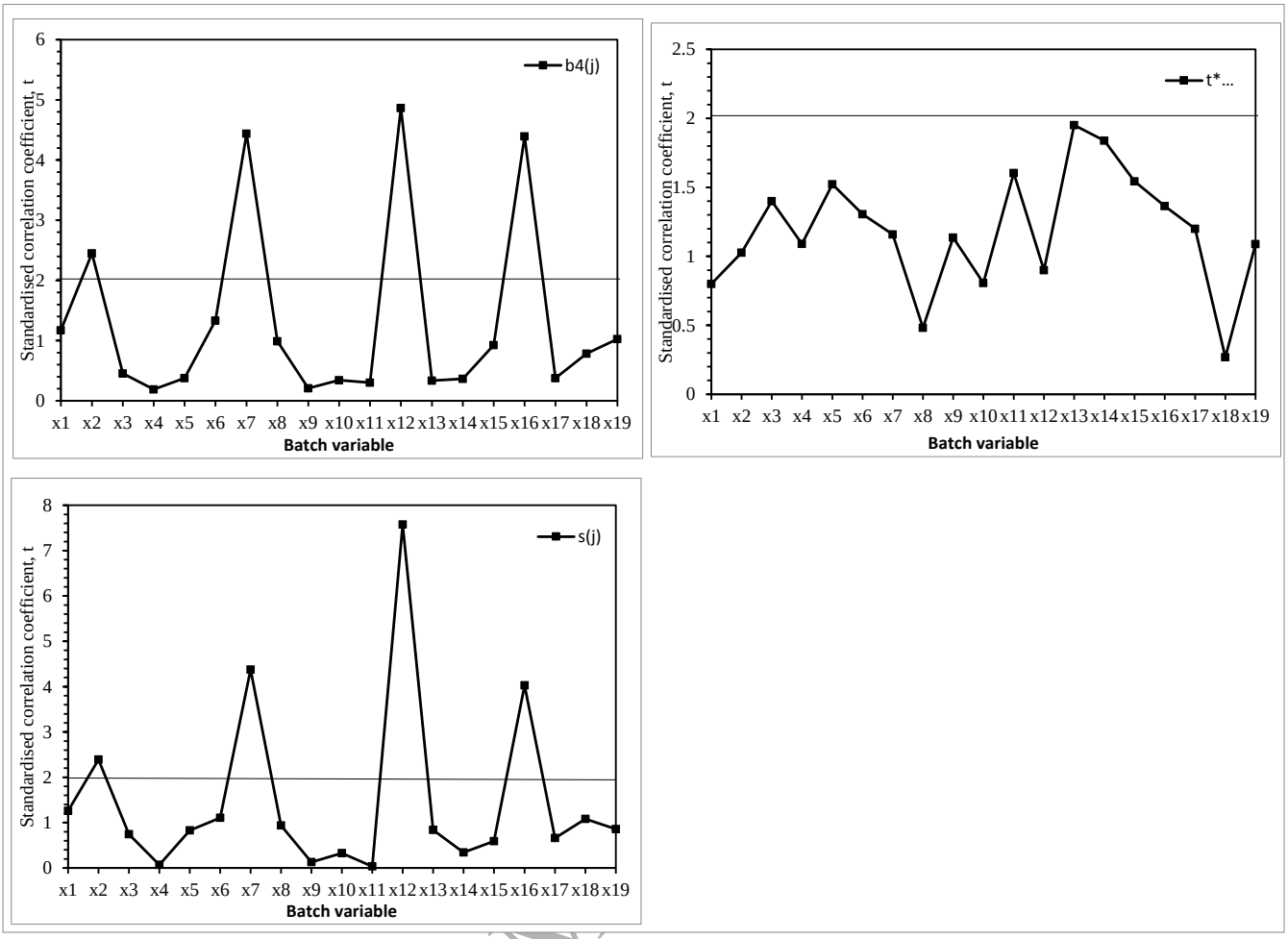
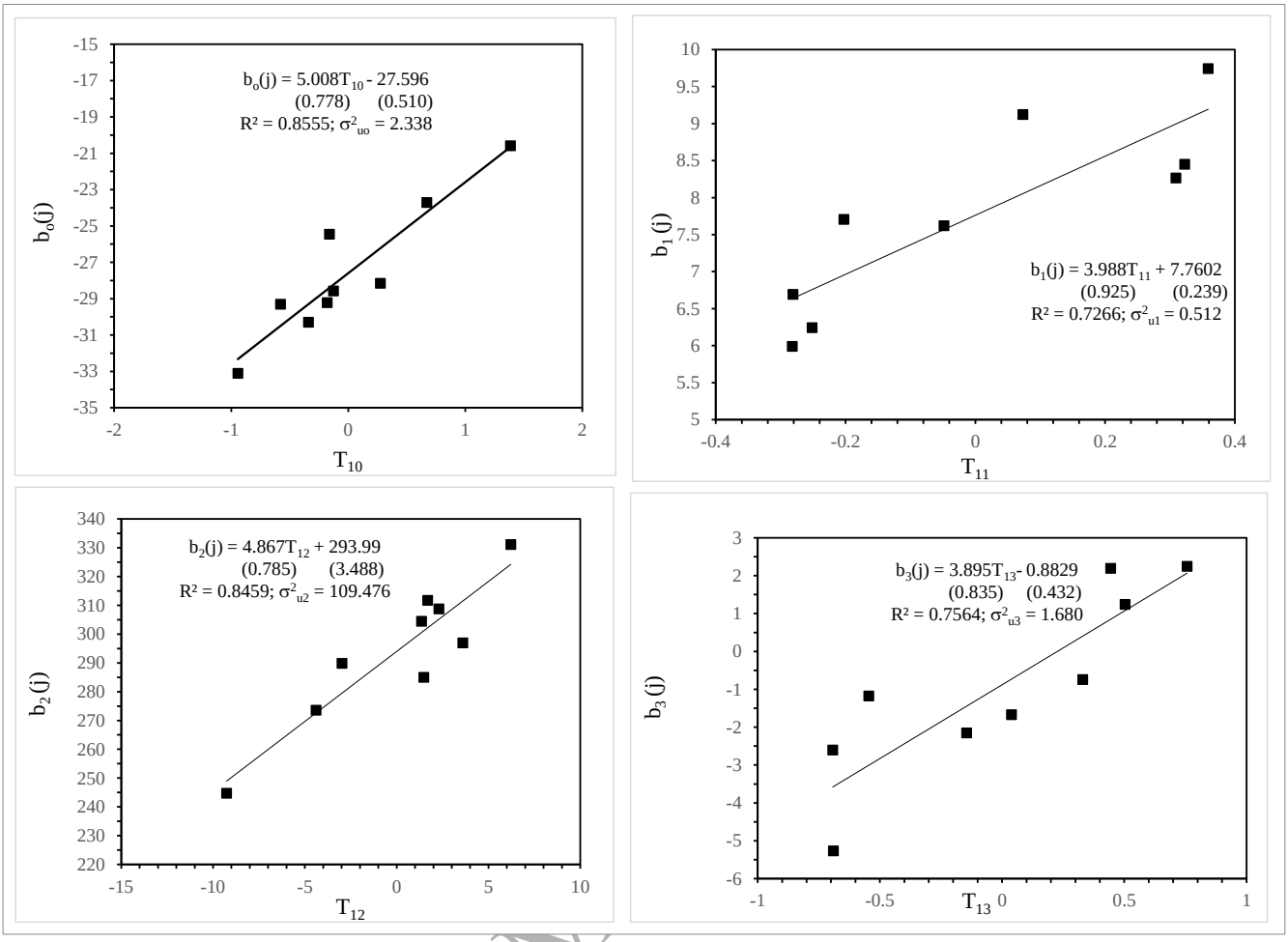


Fig. 3. Plot of the standardised correlation coefficients (given by Eq. (11)) between each Wilshire parameter and each batch variable.



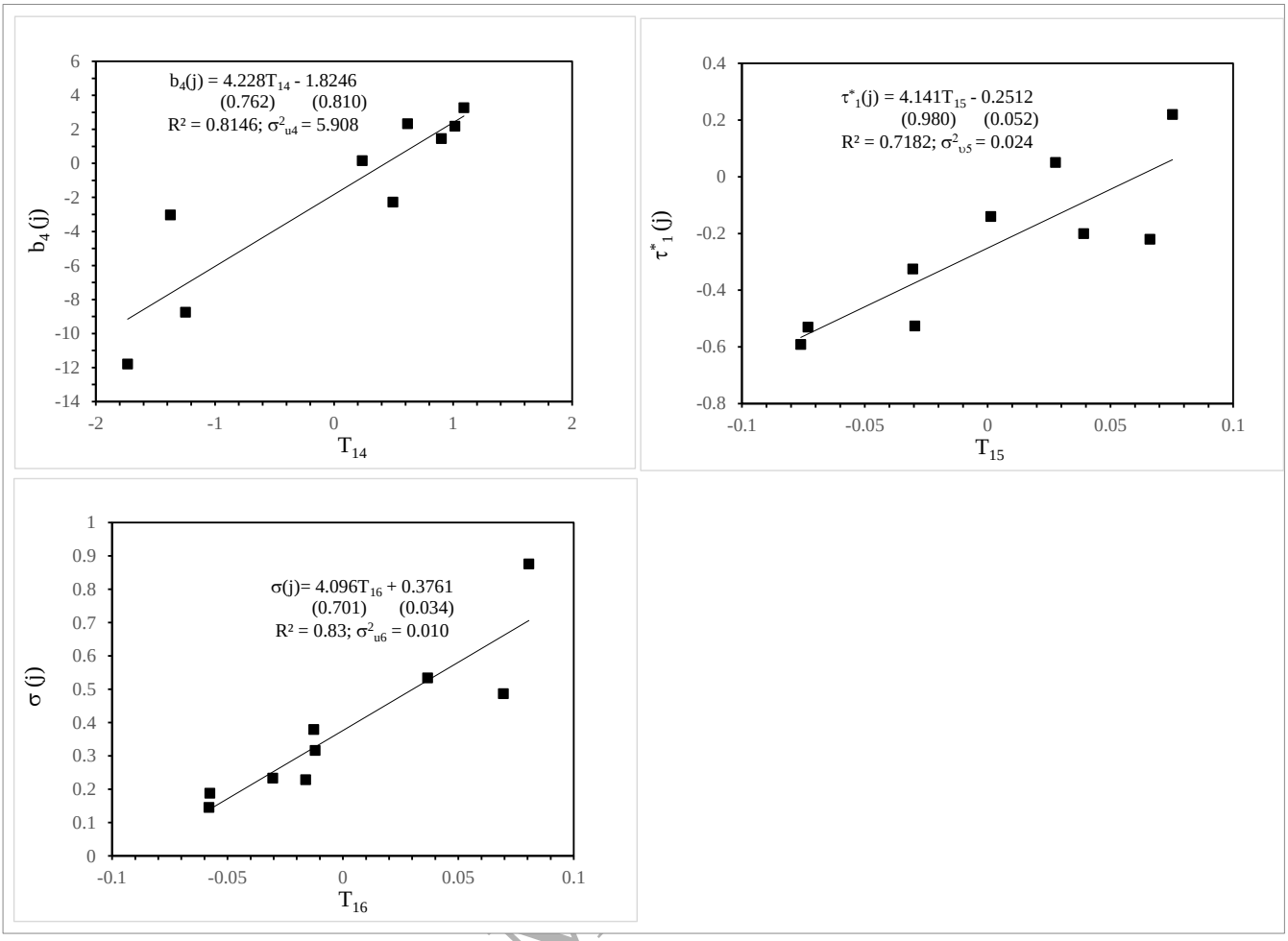


Fig. 4. Plot of the parameters of the Wilshire equation for each batch against their partial least squares components.

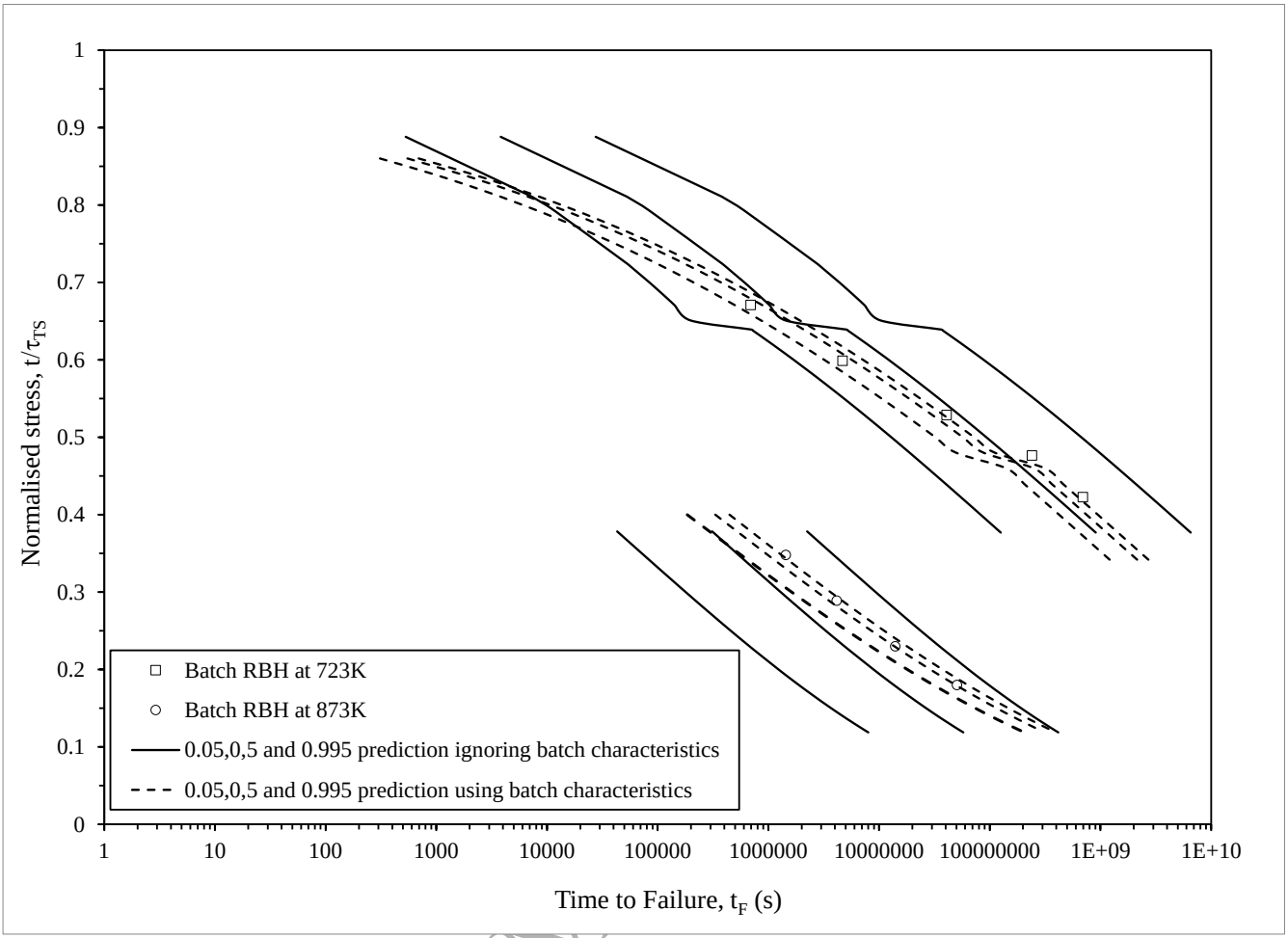


Fig.5a. Plot of times to failure against the normalised stress at 873K and 723K for batch RBH of 12Cr stainless steel, together with the percentile predictions obtained by a. ignoring batch differences (solid curves) and b. accounting for batch differences (dashed curves).

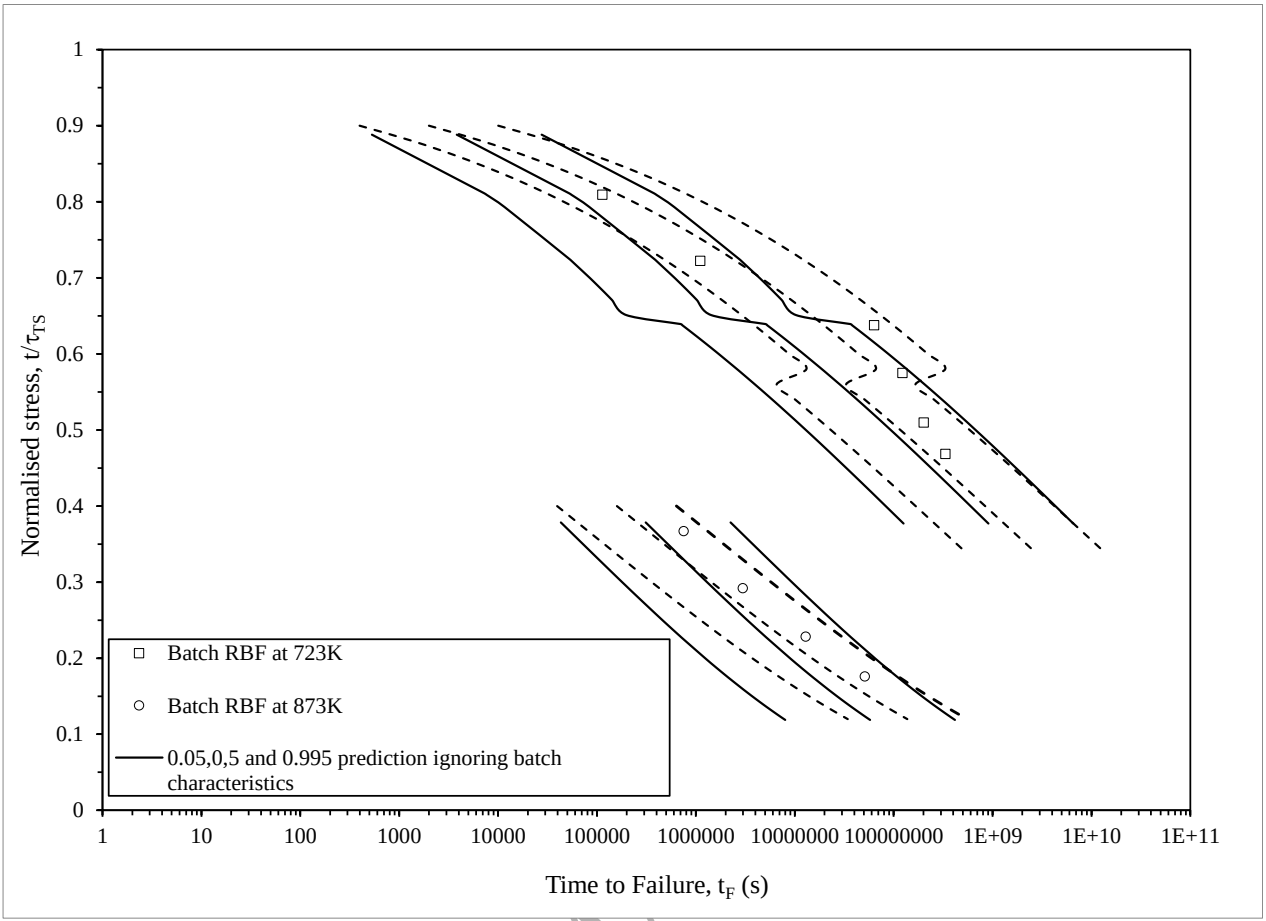


Fig.5b. Plot of times to failure against the normalised stress at 873K and 723K for batch RBF of 12Cr stainless steel, together with the percentile predictions obtained by a. ignoring batch differences (solid curves) and b. accounting for batch differences (dashed curves).

Table 1a

Chemical composition of each batch of 12Cr stainless steel (wt%).

Batch	C	Si	Mn	P	S	Ni	Cr	Mo	Cu	Al	N
RBA	0.12	0.43	0.42	0.022	0.004	0.19	11.8	0.05	0.05	0.023	0.0149
RBB	0.12	0.41	0.46	0.02	0.004	0.23	12.1	0.04	0.06	0.032	0.0137
RBC	0.14	0.31	0.46	0.019	0.008	0.15	12	0.04	0.05	0.008	0.0167
RBD	0.14	0.5	0.67	0.027	0.01	0.32	11.78	0.18	0.21	0.036	0.0175
RBE	0.12	0.48	0.71	0.026	0.007	0.31	12.38	0.09	0.09	0.038	0.0194
RBF	0.12	0.21	0.66	0.028	0.003	0.41	11.8	0.13	0.1	0.002	0.0393
RBG	0.13	0.5	0.61	0.016	0.014	0.43	11.64	0.18	0.08	0.044	0.0264
RBH	0.13	0.36	0.65	0.015	0.011	0.38	11.69	0.19	0.03	0.016	0.027
RBJ	0.14	0.28	0.59	0.018	0.017	0.45	11.92	0.21	0.04	0.007	0.0303

Table 1b

Other batch characteristics of 12Cr stainless steel.

Batch	Austenite grain size	Rockwell Hardness	% non metallic inclusion	Heat treatment
RBA	5.8	98	0.02	1
RBB	5.6	99	0.02	1
RBC	5.2	98	0.02	1
RBD	6.9	94	0.03	2
RBE	7	90	0.04	2
RBF	5.6	93	0.04	2
RBG	7.8	99	0.04	3
RBH	4.8	99	0.05	4
RBJ	5.1	98	0.06	5

OC – Oil cooled. AC = Air cooled.

1 - Forged. 950^0C for 1 hour then OC. 650^0C for 2 hours then AC.

2 - Hot rolled. 970^0C for 0.5 hours then OC. 650^0C for 2 hours then AC.

3 - Forged. 980^0C for 0.5 hours then OC. 650^0C for 2 hours then AC. 630^0C for 2 hours then AC.

4 - Forged. 980^0C for 0.5 hours then OC. 640^0C for 2 hours then AC. 630^0C for 2 hours then AC.

5 - Forged. 980^0C for 0.5 hours then OC. 630^0C for 2 hours then AC. 610^0C for 2 hours then AC.

Table 2
Maximum likelihood estimates of Wilshire parameters: batch by batch.

Parameter/ Batch	$\hat{b}_o(j)$	$\hat{b}_1(j)$	$\hat{b}_2(j)$	$\hat{b}_3(j)$	$\hat{b}_4(j)$	$\hat{\sigma}(j)$	$\tau_1^*(j)$	K	ln[L(j)]
RBA: j=1	-25.4474	9.7443	284.938	-2.6049	-2.2728	0.3164	-0.220	∞	-8.1714
	[-15.30]*	[16.30]*	[27.00]*	[3.95]*	[-1.31]	[6.53]*	[-]		
RBB: j=2	-29.2104	8.4526	304.453	-5.2704	2.3344	0.3790	0.220	12	-14.8987
	[-18.00]*	[22.40]*	[28.00]*	[-5.60]*	[-1.03]	[7.64]*	[-]		
RBC: j=3	-30.2921	8.2671	308.714	-1.1760	1.4576	0.2328	-0.200	∞	1.0193
	[-27.00]*	[23.50]*	[41.50]*	[-3.1]*	[1.38]	[7.22]*	[-]		
RBD: j=4	-28.5751	7.7055	311.727	-0.7452	-11.794	0.8757	-0.526	∞	-34.6494
	[-6.47]*	[7.53]*	[11.1]*	[-0.61]*	[-2.8]#	[7.14]*	[-]		
RBE: j=5	-29.3016	5.9901	296.935	2.1905	-3.0257	0.4861	-0.140	∞	-19.4692
	[-13.70]*	[21.60]*	[21.60]*	[3.95]*	[-1.65]&	[7.51]*	[-]		
RBF: j=6	-33.0998	6.6944	331.079	2.2468	-8.7463	0.5339	-0.591	∞	-19.7668
	[-11.50]*	[10.30]*	[18.30]*	[32.8]*	[-3.47]*	[7.08]*	[-]		
RBG: j=7	-20.5856	7.6208	244.690	-2.1534	0.1626	0.2285	0.051	4	0.4768
	[-17.50]*	[22.70]*	[31.80]*	[-5.48]*	[0.16]	[7.46]*	[-]		
RBH: j=8	-23.7031	9.1229	273.542	-1.6707	2.1863	0.1449	-0.325	1	6.8436
	[-25.70]*	[26.10]*	[43.90]*	[-4.64]*	[2.57]#	[6.06]*	[-]		
RBJ: j=9	-28.1523	6.2439	289.846	1.2373	3.2766	0.1876	-0.530	∞	3.6243
	[-28.20]*	[10.70]*	[44.10]*	[2.13]#	[2.70]#	[6.64]*	[-]		

* Rejects the null hypothesis that the parameter equals zero at the 1% significance level.
# Rejects the null hypothesis that the parameter equals zero at the 5% significance level.
& Rejects the null hypothesis that the parameter equals zero at the 10% significance level
Student t test statistic for the above null hypothesis is shown in parenthesis. [-] means t value not available due to a grid search optimization used to identify τ^*_1.
Ln[L(j)] is the log likelihood for batch j as defined by Eq.(5d)

Table 3
The loadings in partial least squares components for each Wilshire parameter.

Batch variables/ Wilshire parameters	$\hat{b}_o(j)$	$\hat{b}_1(j)$	$\hat{b}_2(j)$	$\hat{b}_3(j)$	$\hat{b}_4(j)$	$\tau_1^*(j)$	$\hat{\sigma}(j)$
x_1	1.2580	-0.3609	-8.5050	0.1241	-2.1013	0.0781	0.0968
x_2	0.6305	0.2508	-3.9342	-0.5478	1.0931	0.0302	-0.0469
x_3	49.819	-49.2047	-485.364	99.3708	62.8566	-9.0240	-4.3619
x_4	35.4676	-13.1331	-190.545	27.1627	41.0227	-11.2677	-0.6271
x_5	19.9344	2.2065	-117.957	-8.7688	-7.1979	1.3214	0.6630
x_6	-1.0842	-7.6955	4.5847	15.5683	-22.2161	-1.1346	0.8266
x_7	-541.691	-103.967	3844.799	248.2894	-941.769	-22.8802	40.7797
x_8	401.4471	-76.6911	-3005.49	81.9938	385.2947	-10.2645	-16.0234
x_9	8.2209	-6.4985	-64.9286	10.6465	-3.8289	-0.9958	-0.1019
x_{10}	-8.4554	-2.4034	40.0002	2.2238	2.9513	0.3465	0.1229
x_{11}	19.9715	-6.3173	-123.632	12.9327	-8.6641	-2.0444	0.0428
x_{12}	-15.9610	-6.0024	164.0345	9.0344	-85.2408	-1.6236	3.9829
x_{13}	122.9791	2.9370	-771.993	-57.2438	-43.4733	10.6142	4.5261
X_{14}	-48.6007	-73.1194	172.0446	178.598	-84.9226	-18.3903	-3.4670
X_{15}	-1.0804	1.5917	8.0652	-3.2013	3.4965	0.2768	0.1001
X_{16}	-4.0937	-1.4453	28.8832	3.1704	-9.0461	-0.2517	0.3837
X_{17}	7.8871	-0.1568	-55.4643	-1.4293	2.2356	0.34	-0.1661
X_{18}	4.3799	1.5331	-23.0058	-0.8863	4.5123	-0.083	0.2601
X_{19}	-0.6254	-1.7058	-4.6638	2.3852	5.7388	-0.3136	-0.2121

x_1 = Austenitic grain size; x_2 = Rockwell hardness; x_3 = % non metallic inclusions; x_4 = C; x_5 = Si; x_6 = Mn; x_7 = P; X_8 = S; x_9 = Ni, x_{10} = Cr; x_{11} = Mo; x_{12}= Cu; x_{13} = Al; x_{14} = N; x_{15} = Heat treatment 1; x_{16} = Heat treatment 2; x_{17} = Heat treatment 3; x_{18} = Heat treatment 4; x_{19} = Heat treatment 5.
All chemical elements measured in % weight.
Heat treatment variables are dummy variables taking on a value of 1 when that treatment applies and zero otherwise.
All shown loading are the d1kq values of Eq. (A1)