Prediction of Pressure Tube Fracture Toughness using a Multivariate Regression Model

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Abstract

The fracture toughness of the zirconium alloy (Zr-2.5Nb) is an important parameter in determining the flaw tolerance for operation of pressure tubes in reactor. Fracture toughness data have been generated by performing rising pressure burst tests on sections of pressure tubes removed from operating reactors. The test data were used to generate a lower-bound fracture toughness curve, which is used in defining the operational limits of pressure tubes. The paper presents a comprehensive statistical analysis of burst test data and develops a multivariate statistical model to relate toughness with material chemistry, mechanical properties, and operational history. The proposed model can be useful in predicting fracture toughness of specific in-service pressure tubes, thereby minimizing conservatism associated with a generic lower-bound approach.

1. Introduction

The pressure tubes used in CANDU® reactors are fabricated from cold-worked Zr-2.5Nb and have a length of 6.3 m, inside diameter of 103 mm, and a wall thickness of 4.2 mm. During service, irradiation and deuterium ingress from the pressurized heavy water coolant reduces the fracture toughness of the pressure tube material. Periodic assessments of surveillance tubes removed from reactors are conducted to ensure that the tubes remain "fit-for-service" [1]. An important parameter in determining the flaw tolerance for operation of pressure tubes, and predicting "leak-before-break" scenarios, is the critical crack length (CCL). Measurements of the CCL were initially obtained from rising pressure, burst tests on sections of pressure tubes removed from operating reactors [2]. In these early burst tests, tubes with different lengths of through-wall axial cracks were pressurized to failure. The CCL was taken as the initial crack length associated with failure for a given hoop stress [2]. An alternative method based on the measurement of a J-resistance (J-R) curve to determine a critical stress intensity factor (K_c) has been developed at AECL [3]. In this approach, the results from individual burst tests are used to measure the resistance to crack growth to the point of instability, for a given test temperature. The stress intensity at the point of instability is then calculated, and taken to be K_c for the given conditions. Overall, this standardised method is more cost effective, and less wasteful of material. As a result, the standardized burst test, in combination with small-scale specimens, has been used to investigate the factors influencing the toughness of irradiated pressure tube material [4,].

To-date, 106 burst tests have been performed on sections of 33 irradiated Zr 2.5Nb pressure tubes removed from operating reactors using the standardised method [3]. The measured values of K_c from a portion of these tests were used to generate a lower-bound curve, thereby defining the operational limits of pressure tubes. Such a conservative approach was deemed necessary due in part to the significant tube-to-tube variability in measured fracture properties. Previous studies have identified specific material characteristics that influence pressure tube fracture toughness, and the variability in the burst test results. The role of chlorine in the formation of primary void nucleation sites for fracture, for example, highlighted the importance of controlling the chemical composition and fabrication routes of pressure tubes [3, 5].

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In the current study, a comprehensive multivariate statistical analysis of the burst test database is performed to correlate fracture toughness with relevant covariates, such as material chemistry, mechanical properties and irradiation history. As a result, a significant portion of the burst test fracture toughness variability is addressed, and the statistical influences of the covariates are quantified. As part of the analysis, predictive models for pressure tube fracture toughness are developed from the most significant covariates. The outline of this paper is as follows. First, a description of the irradiated Zr-2.5Nb pressure tube material and burst test methodology is presented. Next, the data used in the statistical calculations are summarised, followed by a description of the multivariate statistical techniques used in the study. The results from the multivariate analysis are then presented, followed by concluding remarks.

2. Material

A total of 106 burst tests have been performed on irradiated specimens (~ 0.5 m long) taken from 33 Zr-2.5Nb pressure tubes. The majority of specimens initially tested were from sections of tubes removed after approximately 18 years of operation, and irradiation to a fast neutron fluence of up to 11×10^{25} n m^{-2} (E > 1 MeV) at temperatures in the range 249 °C (inlet end) to 290 °C (outlet end). These tubes were fabricated as standard cold-worked (~ 26%) Zr-2.5Nb pressure tubes prior to 1987, and it was specified that the ingots should be vacuum arc melted twice, as this process reduces some of the volatile impurity elements [6]. Some ingots, however, were produced using 100% recycled material, which is equivalent to the ingot being melted four times. The multiple melting of the material significantly reduced some of the volatile impurity elements (e.g. chlorine), which has a significant effect on the fracture toughness [5].

Cold-worked Zr-2.5Nb pressure tubes manufactured prior to 1987 were fabricated in accordance with the chemical specifications detailed in [6], which do not include any specific limits on the concentrations of impurity elements such as chlorine and phosphorus. Previous studies have demonstrated that these particular elements are among a few that have a significant effect on the deformation and fracture behaviour of pressure tube material [3, 5]. As a result, changes to the chemical composition specifications for Zr-2.5Nb pressure tubes were recommended [7] to improve the properties of newer tubes. Although the manufacturing route for pressure tubes has evolved with time, the overall changes to fabrication have not been substantial.

More recently, burst tests have been performed on material having maximum fluences in the range 5.5×10^{25} n m^{-2} to 18.1×10^{25} n m^{-2} (E > 1 MeV), and irradiation temperatures ranging from 249 °C to 293 °C. It is noted that all of the pressure tube sections used in the burst test program had total hydrogen equivalent concentrations of hydrogen isotopes that were less than 30 ppm by weight (0.27at %)[1].

The chemical compositions of the 33 tubes used in the burst test program were taken from ingot analyses provided by the manufacturer, and Glow Discharge Mass Spectrography (GDMS) of offcuts (material removed from the ends of a pressure tube before installation in reactor). The measured values for the elements chlorine (Cl), carbon (C), oxygen (O), iron (Fe), and phosphorus (P) were used as part of the current study.

3. Burst Test Procedure and Analysis

The standardised procedure for conducting burst tests on irradiated Zr-2.5Nb pressure tube material was developed at AECL [3], and involves spark machining a through-wall axial crack of 55 mm length at the centre of a 0.5 m long section of tube. In addition, results from tests with non-standard

[1] All reference to ppm in text implies ppm by weight.

crack lengths are included in this investigation (initial crack lengths $2a_0$ were in the range of 36.1 mm $\leq 2a_0 \leq$ 86.4 mm). The machined flaw is then sealed with a composite patch made of Teflon, stainless steel, and aluminium sheet that are secured to the pressure tube with silicone rubber. The test section is fitted with mechanical end caps before attachment to the pressurizing system, and enclosed in a protective bell-jar. The machined flaw in the specimen is extended approximately 5 mm axially in each direction by fatigue pressure cycling at room temperature using water and a maximum stress intensity of 15 MPa√m. Stable crack growth is monitored using the direct current potential drop method. Once an experiment is to be conducted, the test section is heated to the desired test temperature using external heating coils and held for at least one hour. The test section is then pressurized with argon gas monotonically until failure. The Dugdale strip yield equation for an axial, through wall defect in a pressurized cylinder is used to calculate the Mode I stress intensity factor (K_c) as [3],

$$K_I = \left(\frac{8\sigma_f^2}{\pi} a \ln \left[\sec \left(\frac{\pi M \sigma_h}{2\sigma_f} \right) \right] \right)^{1/2}, \tag{1}$$

where σ_f = flow stress (mean of the yield stress and ultimate tensile strength), $2a$ = total crack length, σ_h = hoop stress (pr_i / t), p = internal pressure, r_i = internal radius, t = wall thickness, and M = Folias bulging correction factor given approximately by [8]:

$$M = \{1 + 1.255[a^2 /(r_m t) - 0.0135[a^4 /(r_m t)^2]\}^{1/2}, \tag{2}$$

for a given mean radius r_m.

The resulting fracture toughness is expressed as the critical stress intensity factor K_c, corresponding to the stress intensity at the point of instability (rupture) calculated using the initial crack length $2a_0$ rather than the crack length at the point of instability ($2a_i$). As a result, K_c represents a conservative estimate of the fracture toughness. The CCL determined from K_c is also conservative provided that the pressure at rupture is less than the operating pressure.

4. Statistical Data Analysis

4.1 Preliminary Analysis

The database for statistical analysis consists of fracture toughness (K_c) values obtained from 106 tests and values of 12 covariates listed in Table 1 for each test sample. The average and standard deviation of K_c are estimated as 113.55 MPa√m and 32.83 MPa√m, respectively, and the coefficient of variation is 29 %.

The normal probability paper plot, presented in Figure 1, shows that the normal distribution can model the test data reasonably well. From the fitted distribution, the 10% probability lower bound for K_c is estimated as 71.5 MPa√m. It should be remarked that the lower and upper tail regions of the empirical distribution (i.e., data) are not well represented by the normal distribution, and the use of other distributions for improving the goodness-of-fit will be explored the future work.

The traditional single-variate probabilistic analysis generally provides a conservative lower bound, especially when the data exhibit large variability. To improve the lower-bound estimate, a regression analysis is required as it can reduce the prediction variability by establishing a statistical relation with other random variables (covariates) that influence the fracture toughness.

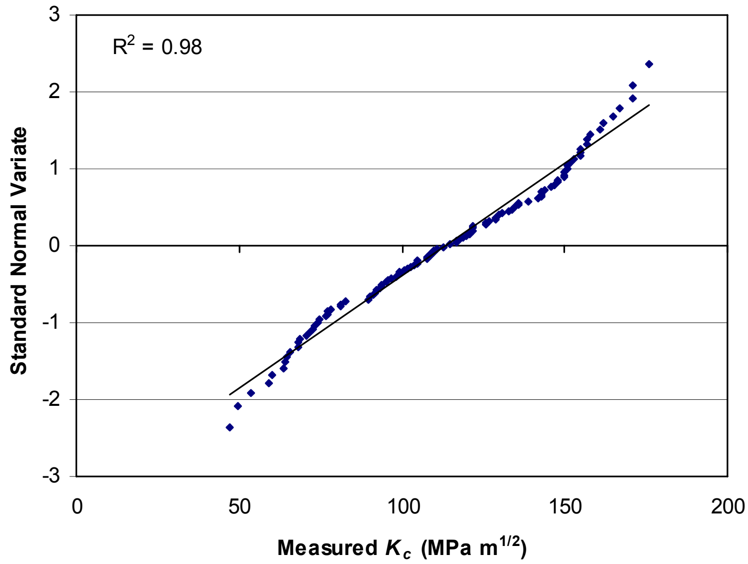

Figure 1: Normal probability plot of fracture toughness data

Table 1: Variables affecting the fracture toughness

Variables		Name	Mean	Standard Deviation	Units
Material Chemistry	X_1	Chlorine [C1]	4.312	3.775	ppm
	X_2	Phosphorous[P]	25.033	17.781	ppm
	X_3	Carbon [C]	159.948	25.971	ppm
	X_4	Oxygen [O]	1133.50 9	95.491	ppm
	X_5	Iron [Fe]	756.321	259.527	ppm
Mechanical Property	X_6	Flow stress	922.330	146.219	MPa
Operational Parameters	X_7	Irradiation Fluence	9.435	2.188	10^{25}n/m^2
	X_8	Irradiation Temperature	266.849	11.148	°C
	X_9	Test Temperature	183.830	90.793	°C
Material Texture Parameters	X_{10}	OFFCUT Avg Fr^*	0.322	0.0243	
	X_{11}	OFFCUT Avg Ft^*	0.628	0.0268	
	X_{12}	OFFCUT Avg Fl^*	0.0495	0.00977	

* *Fr*, *Ft*, and *Fl* are measures of the fraction of grains with basal plane normals oriented in the radial, transverse, and longitudinal tube directions, respectively.

The dependence of K_c on some key variables or covariates is illustrated in Figures 2 and 3. For a given test temperature, K_c exhibits a significant variability, as seen from Figure 2(a). The average fracture toughness increases with increasing test temperature, to ~150 °C, and then it is approximately constant. The chlorine concentration has more pronounced influence, as seen from Figure 2(b). Here, K_c decreases as the chlorine concentration increases. The effects of irradiation fluence and temperature are more variable as no definite trends are apparent from data plotted in Figure 3. The effects of other covariates can be studied similarly.

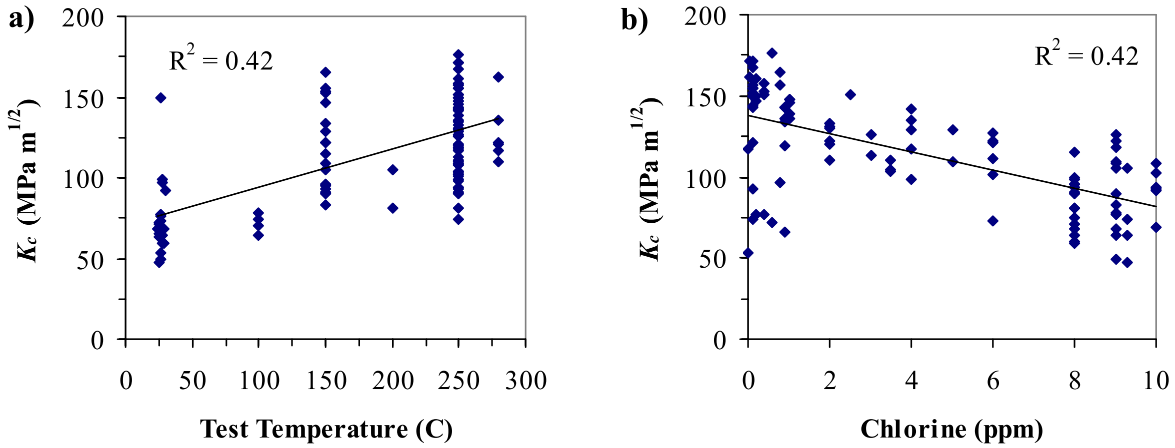

Figure 2: Measured K_c as a function of (a) test temperature, (b) chlorine concentration

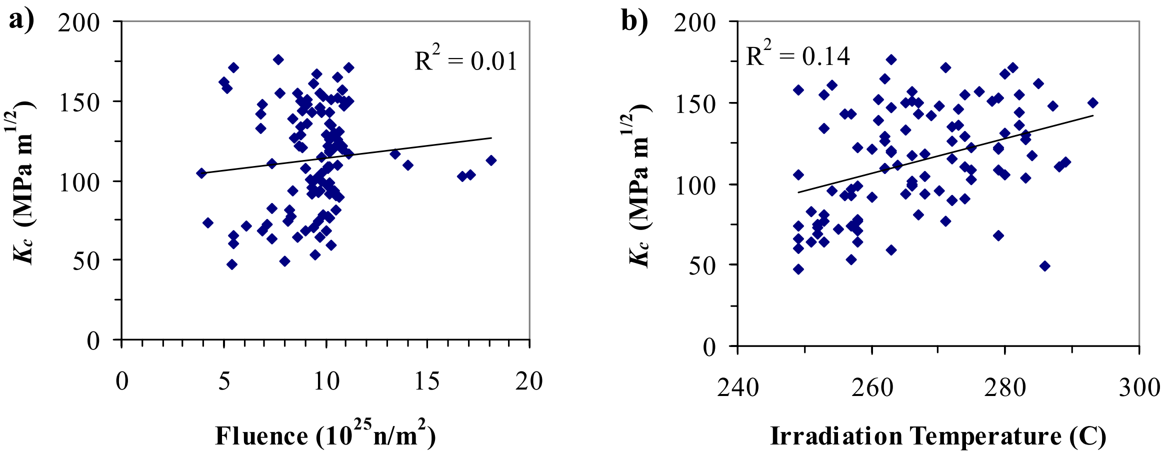

Figure 3: Measured K_c as a function of (a) irradiation fluence, (b) irradiation temperature

4.2 Multivariate Regression Analysis

The variability associated with fracture toughness cannot be explained completely by any single parameter. Therefore, a multivariate regression analysis is required for data analysis and model building purposes.

The basic premise of the regression analysis is to model the fracture toughness (Y) as a random variable in the following way:

$$Y = \hat{Y} + E\,, \tag{3}$$

where $\hat{Y}$ is the mean of Y and E denotes a normally distributed random error, also referred to as the residual. The average of the fracture toughness is related with 12 covariates (**X**) through a linear regression equation,

$$\hat{Y} = b_0 + b_1 X_1 + b_2 X_2 + \ldots + b_{12} X_{12}\,. \tag{4}$$

The model coefficients (b_k) are determined from the principle of least-squares applied to the available data. The calibrated model can be used for predicting fracture toughness (Y_m) corresponding to a given vector of covariates $\mathbf{x_m}$. The average of Y_m can be calculated from Eq.(4) and the variance associated with it can be evaluated as,

$$s_m^2 = 1 + \{\mathbf{x_m}\}^{\mathbf{T}}[\mathbf{P^T P}]^{-1}\{\mathbf{x_m}\}, \tag{5}$$

where matrix **P** is estimated from available data used in the regression analysis [9]. A lower prediction limit corresponding to a probability level (α), denoted as Y_α, can be used as a probabilistic lower-bound for the fracture toughness of a specific pressure tube with covariates $\mathbf{x_m}$. Thus, the proposed model can account for tube-to-tube variability in a more systematic manner.

Since covariates have different physical units, the regression is carried out after normalizing or standardizing the covariates as $(X_k - \mu_k)/\sigma_k$, where μ_k and σ_k are the mean and standard deviation of a covariate X_k. The magnitudes of coefficients of regression in this standardized space are indicative of their relative importance in explaining the variability associated with the fracture toughness [10]. These standardized regression coefficients should be bounded within ±1, otherwise they are considered inconsistent due to large inter-correlation among covariates.

A general assumption of traditional multiple regression analysis is that all covariates are independent. However, if this assumption is violated due to strong inter-correlation among covariates, the analysis can lead to irrational regression results. To overcome this difficulty, an advanced principal component regression (PCR) method is applied for data analysis [9]. In the PCR method, the original covariates are transformed into principal components, which are independent of each other. The orthogonality of the principal components eliminates the problem of inter-correlation among original covariates. A multiple regression model is fitted to these uncorrelated principal components. The order of the model is selected on the basis of a suitable criterion, such as the proportion of variance explained by the model as indicated by R^2. The most representative model is then transformed back to the original variable space and then used for prediction purposes.

4.3 Results

In the data analysis, eigen-values and eigen-vectors of the covariance matrix (12×12), estimated from the test data, were computed. Since some covariates were strongly inter-correlated, a smaller number of (m) principal components can be sufficient to capture the variability associated with the response variable (Y). The relative magnitudes of eigen-values of the covariance matrix are indicative of their significance in explaining the variability of Y.

The regression model was fitted by varying the number of principal components (m) from 3 to 12, and the goodness of fit in each case was evaluated in terms of R^2. Figure 4 shows that for m>7, R^2 is almost constant at a value of 0.75. Thus, seven principal components are sufficient for building the statistical regression model, and it was derived as:

$$\begin{aligned} y = & -5.5308 - 3.7733X_1 + 0.2258X_2 + 0.0380X_3 + 0.0392X_4 - 0.0040X_5 - 0.0634X_6 \\ & - 3.1136X_7 + 0.6005X_8 + 0.1009X_9 + 10.9552X_{10} - 1.8062X_{11} - 222.6954X_{12} \end{aligned} \tag{6}$$

This model explains 75% of the variability in the data, i.e., sum of squares of deviation from the mean of K_c data. Another measure of model quality is the magnitude of standard error in relation to the standard deviation of the data. The standard error of the model with 93 degrees of freedom is 17.38 MPa√m, which is approximately half of the standard deviation of the average K_c (32.83 MPa√m). In other words, the regression model reduces the standard deviation associated with the original data by 47%.

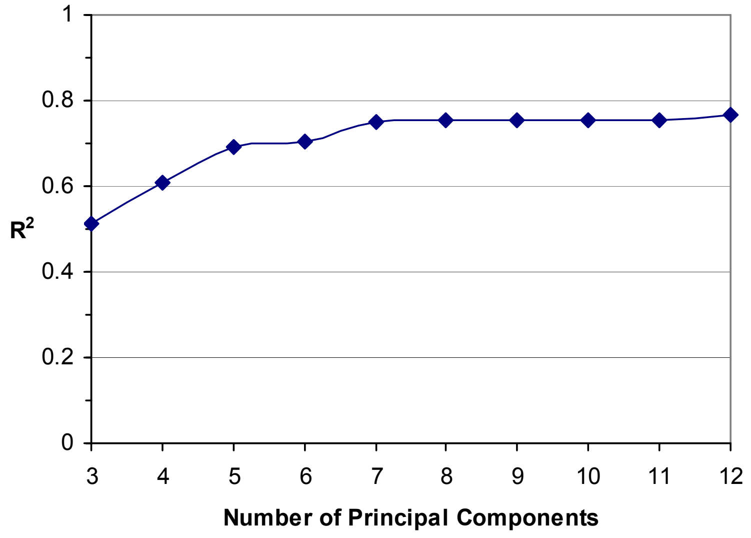

Figure 4: Variation of R^2 with the number of principal components

The values predicted from the regression model are compared with actual K_c data in Figure 5. The residuals, i.e., the differences between measured and predicted values, are plotted on a normal probability plot in Figure 6. It shows that the residuals can be fitted quite closely by a normal distribution, which validates the modelling assumption underlying the regression analysis.

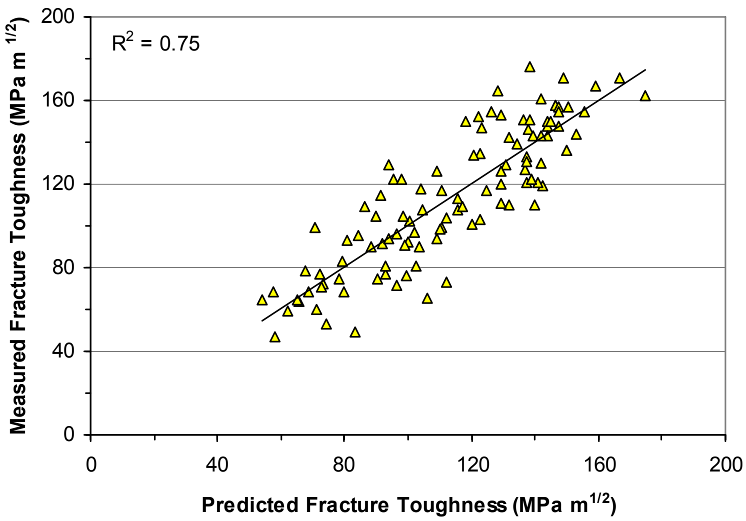

Figure 5: Comparison of regression model predictions with measured values of K_c

The importance and nature of the effect of a covariate on K_c can be investigated by examining the sign and magnitude of standardized regression coefficients (Figure 7). The negative sign indicates that increasing the value of that covariate would decrease the toughness, and the reverse is implied by the positive sign. It shows that chlorine concentration has the highest influence on the fracture toughness, followed by flow stress, test temperature, irradiation fluence, and irradiation temperature. Further investigations into the inter-correlation of the covariates, and the functional relation between individual covariates and fracture toughness are being addressed in ongoing work.

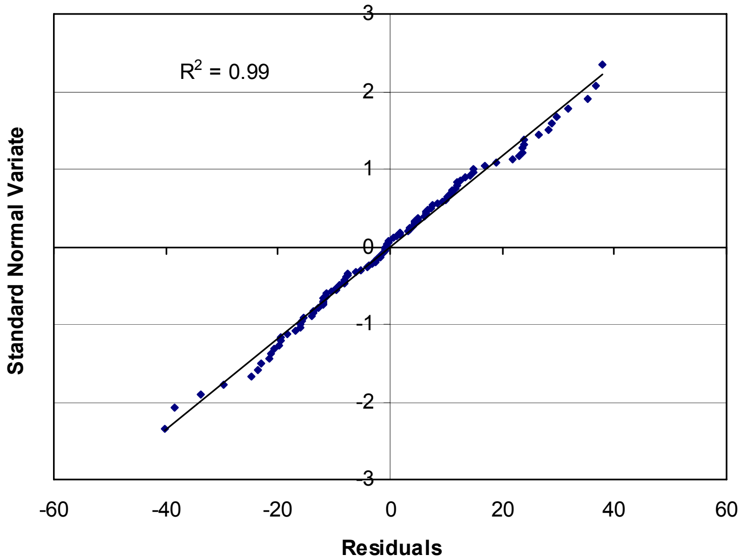

Figure 6: Normal probability paper plot of residuals for model validation

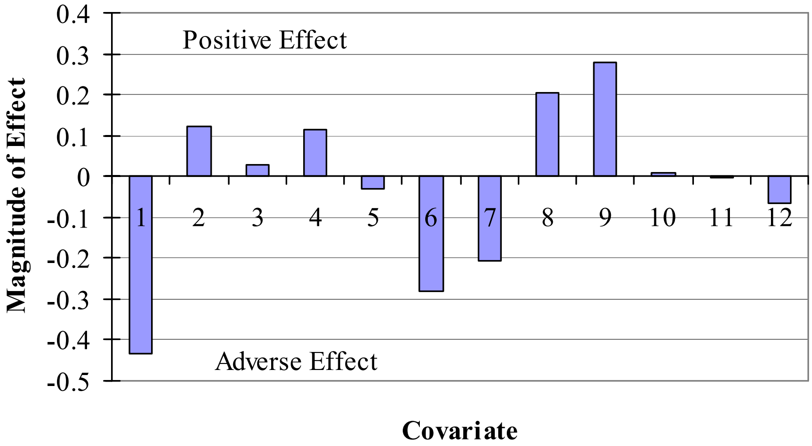

Figure 7: Effect of covariates on the fracture toughness (variable numbers from Table 1)

4.4 Predictive Model for In-Service Pressure Tubes

In many cases, information on all 12 covariates (Table 1) for a given in-service pressure tube may not be available. Therefore, the predictive model has to be constructed using a subset of variables. Recognizing that material chemistry and operational conditions can easily be obtained for all in-service pressure tubes, a regression model is developed using only 5 covariates, namely, chlorine (X_1), oxygen (X_4), irradiation fluence (X_7), irradiation temperature (X_8) and test temperature (X_9). The model equation from ordinary multiple regression analysis is estimated as,

$$y = -92.203 - 4.287X_1 + 0.05X_4 - 2.869X_7 + 0.595X_8 + 0.194X_9 \,. \qquad (7)$$

As the correlations among these five variables are small, the principal component regression method was not required. A comparison of the observed and predicted values is shown in Figure 8. It is notable that the R^2 value for this model (0.75) is essentially the same as the full model involving all 12 covariates. The standard error of the model with 100 degrees of freedom is 16.90 MPa√m, which is slightly less than that of the full model given in Eq.(6). The normal probability paper plot of residuals in Figure 9 confirms the validity of the assumption of the normality of the model error. These results imply that the additional covariates used in the previous analysis (Section 4.3) may not contribute significantly to fracture toughness. Future work will address this issue.

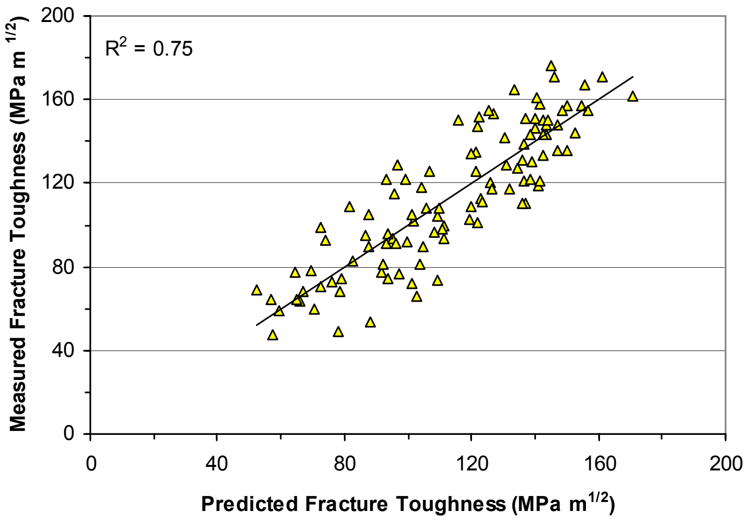

Figure 8: Regression model for use for in-service pressure tubes

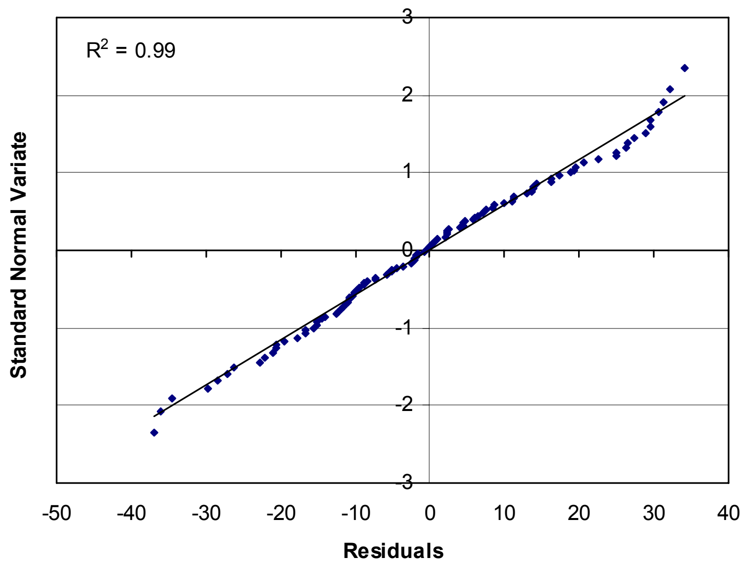

Figure 9: Residuals of the model in Eq.(7) plotted on the normal probability paper

The effects of the covariates on K_c for this analysis are presented in Figure 10. Again, a negative sign implies that increasing the value of the covariate results in a decrease in the toughness (adverse effect), and a positive value implies a positive affect. From Figure 10, it is seen that relative influence of the individual covariates is similar to that observed in the previous analysis (Figure 7). However, in this analysis test temperature has the highest influence (positive), followed by [Cl] (adverse), irradiation temperature (positive), irradiation fluence (adverse), and oxygen (positive). It is worth noting that other elements could have been chosen (e.g. phosphorus rather than, or in addition to oxygen), but the objective was to determine the overall effect of reducing the number of covariates. Future work will be performed to optimise a model for in-service pressure tubes.

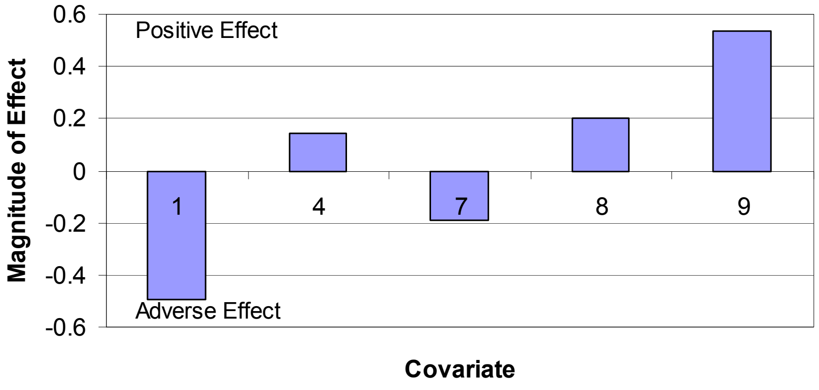

Figure 10: Effect of covariates on the fracture toughness for in-service pressure tube analysis (variable number from Table 1)

5. Conclusions

A multivariate statistical analysis of the Zr-2.5Nb pressure tube burst test fracture toughness database was performed as a first step to correlate all relevant covariates, such as material chemistry, mechanical properties and irradiation history. The analysis was based on an advanced Principal Component Regression (PCR) method, and showed that a significant portion (~ 75%) of the burst test fracture toughness variability can be addressed. The PCR approach is superior to ordinary least squares regression, as it accounts for correlation among the covariates. A predictive model, that correlates 12 variables (listed in Table 1) with pressure tube fracture toughness, was developed and the relative importance of the variables quantified. An additional model was developed consisting of 5 covariates that are readily available for in-service pressure tubes (chlorine concentration, oxygen concentration, irradiation fluence, irradiation temperature, and test temperature). Results indicated that this reduced model had approximately the same predictive capability as the full (12 covariate) model, thereby demonstrating the importance of individual covariates.

The models developed in the current study demonstrate that multivariate statistical methods can be used to relate pressure tube fracture toughness with covariates such as material composition, mechanical properties and other parameters related to in-service conditions. These models can be used to predict the fracture toughness of in-service pressure tubes, and define the corresponding probabilistic lower-limit based on existing data. These results will improve the understanding of fracture toughness variability for in-service tubes, and may provide a basis for positive (less conservative) changes to guidelines for fitness-for-service assessment.

Acknowledgements

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