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A novel process-linked assembly failure model for adhesively bonded composite structures

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ABSTRACT

The globally growing market for polymer composites and their increasing use within aircraft structures has necessitated reliable bonding of composite laminates to prevent structural failure. However, knowledge behind the interaction between curing process parameters and the failure of polymer composite bonded joints is not keeping pace with the market. A novel nonlinear correlation analysis has been employed and applied to experimental data, to attentively quantify the effect of curing parameters on the failure of bonded composite assemblies. The materials (adherends and adhesive) and the bonding processes were selected from those used in assembly of composite aircraft structures.

1. Introduction

The inevitable process-linked structural performance in adhesively bonded polymer composite structures necessitates an urgent need for reliable, controllable and measurable bonding in composite joint assembly and manufacturing. This need is intensified by the fact that no method of measuring properties prior to installation exist to account for variabilities caused by process control during adhesive bonding, and no non-destructive inspection is available to ensure bond integrity [1,2]. Due to such process-linked performance, certified procedures may not produce reliable bonded assemblies with adequate levels of continuing airworthiness for aircraft structures.

Integrated structural adhesive bonds often present significant technical challenges due to the mismatch in mechanical properties between the bonded members (adherends). Correct bonding and integration require knowledge-based methodology, including structural performance modelling (e.g. see Ref. [3]), that quantifies the effects of each bonding process parameter on the structural response. Existing models for predicting the response of composites have been developed with no or little contribution of such process-linked properties [3]. Those models assume that the curing process has fully been accomplished, or slight effects from incomplete curing. This paper addresses this missing gap and explains the interaction between the curing process in a thermoset polymer bond and its achieved mechanical properties.

A nonlinear correlation analysis approach is used to obtain the level of interaction between the main process parameters and the composite bonded joint's mechanical response. This is a novel employment of this approach that accounts for the process parameters in a simple and straightforward manner based on experimental data. Bond deficiencies are mimicked in single-lap composite bonded joints. Curing process parameters are altered, and their effect on the joints failure is obtained. Finally, the correlation method is applied to quantify the effect of each parameter on the response of the joint. The model is recommended to designers and researchers in academia and industry for understanding and quantification of the effect of process-induced deficiencies in composite assemblies.

2. Nonlinear correlation analysis

Considering a system with multiple inputs and outputs, the Error Reduction Ratio (ERR)-Causality approach [4,5] is a correlation method used to measure the effects of each input parameter on outputs in an interactive system, particularly when the interaction is nonlinear. The effects are quantified in a range from 0% to 100%, the larger the ERR, the higher the dependence between selected input and output. The ERR-Causality approach is underpinned by the nonlinear auto-regressive moving average model with exogenous inputs (NARMAX), detailed in Ref. [4], suitable for a complex system with an unknown inner structure (herein a curing bond). Compared with machine learning methods, one advantage of the NARMAX model is transparency, meaning that it can be written down and therefore easily understood. ERR-Causality has successfully been applied to brain signal analysis, climate change and non-destructive testing [6,7]. However, its application to composite structure manufacturing is novel. This research has focused on understanding and quantification of the interaction between major curing parameters and their resulting bond failure in a critical composite bonded assembly (e.g. aircraft). The process-linked failure is a multi-parameter nonlinear problem. The purpose of the ERR model developed in Refs. [4,5] is to reveal

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any hidden nonlinear interaction. Traditional methods, such as coherence and cross-spectrum, usually assume that the system is linear and stationary, and hence cannot sufficiently reveal and characterise hidden information in a complex system that is nonlinear and dynamic. Moreover, in cases with limited number of tests, applying a statistical analysis cannot be suggested. The ERR is more appropriate and easier-to-implement for laboratory scale tests than the statistical models.

In the ERR model formerly developed in Ref. [5], the composite bonded joint is taken as the system. Curing duration and heating rate in curing bond are taken as the system inputs, and failure load, displacement and strain energy are taken as the system outputs (energy is calculated from 0.5 × load × displacement as the load–displacement curves were linear in our experiments). These inputs are controlled in the experiments. Alternative inputs could have been selected e.g. surface treatment and contamination. However, these inputs are constant in all tests to allow the effects of the curing parameters to be interrogated only. The bond area was degraded in some joints to account for contamination.

2.1. ERR-Causality method

The orthogonal least squares algorithm has been used in the proposed method. This is a popular algorithm used for nonlinear systems. It searches through all possible candidate model terms to select the most effective ones. These are then used to build the model expression [5]. The significance of each selected model term is measured by the ERR index which indicates how much of the change in the system response (output), in percentage, can be accounted for by including the relevant model terms containing inputs. Consider a function with a linear form of terms:

$$y(k) = \sum_{i=0}^{N} \theta_i p_i(k), k = 1, 2, \ldots, M \tag{1}$$

where $y(k)$ is the system output (mechanical response herein) to regress upon. $p_i(k)$ are regressor terms constructed by input variables $\{u_i\}$. θ_i is the vector of unknown coefficients of regressions to be estimated, M denotes the number of data points in the data set, and N denotes the number of terms in the model that is yet to be determined. Eq. (1) can be written as

$$Y = P\Theta \tag{2}$$

where

$$Y = \begin{bmatrix} y(1) \\ y(2) \\ \vdots \\ y(M) \end{bmatrix}, P = \begin{bmatrix} P^T(1) \\ P^T(2) \\ \vdots \\ P^T(M) \end{bmatrix}, \Theta = \begin{bmatrix} \theta(1) \\ \theta(2) \\ \vdots \\ \theta(M) \end{bmatrix} \tag{3}$$

and $P^T(k) = (p_1(k), p_2(k), \ldots, p_N(k))$. Matrix P is decomposed as $P = W \times A$ where

$$W = \begin{bmatrix} w_1(1) & w_2(1) & \ldots & w_N(1) \\ w_1(2) & w_2(2) & \ldots & w_N(2) \\ \vdots & \ddots & \ddots & \vdots \\ w_1(M) & w_2(M) & \ldots & w_N(M) \end{bmatrix}, \tag{4}$$

and $A = \{a_{ij}\}$ is an upper triangular matrix with unity diagonal elements. Therefore, Eq. (2) is re-written as

$$Y = WG \tag{5}$$

where $G = A\Theta = [g_1 \quad g_2 \quad \ldots \quad g_N]^T$. Eq. (5) is now ready to represent the relation between Y and G. We then estimate the effect of each model term to the system output (Y). Values are initially set at $a_{ij} = 0$ for $i \neq j$ (A then becomes an identity matrix), as such $w_1(k) = p_1(k)$. g_1 is calculated from

$$g_1 = \frac{\sum_{k=1}^{M} w_1(k) y(k)}{\sum_{k=1}^{M} w_1{}^2(k)} \tag{6}$$

For $j = 2, 3, \ldots, M$ set $a_{jj} = 1$, thus

$$a_{ij} = \frac{\sum_{k=1}^{M} w_i(k) p_j(k)}{\sum_{k=1}^{M} w_i{}^2(k)} \tag{7}$$

where $i = 1, 2, \ldots, j-1$. The algorithm then calculates

$$w_j(k) = p_j(k) - \sum_{i=1}^{j-1} a_{ij} w_i(k) \tag{8}$$

and

$$g_1 = \frac{\sum_{k=1}^{M} w_j(k) y(k)}{\sum_{k=1}^{M} w_j{}^2(k)} \tag{9}$$

The ERR values for each term p_i is finally defined as

$$ERR_i = \frac{g_1^2 \sum_{k=1}^{M} w_i{}^2(k)}{\sum_{k=1}^{M} y^2(k)} \tag{10}$$

The larger the ERR, the higher dependence between the $\{p_i\}$ terms and the output, Y, an index to indicate the importance of each term (constructed by the process parameters as inputs) for the output, the mechanical response.

3. Composite joints: assembly, materials and processing

Composite single-lap bonded joints are the most common, economic and easily repeatable joints used to measure the performance of adhesively bonded structures. It is the weakest joint configuration as a result of loading eccentricities causing adherend bending which produces high stress concentrations in the through–thickness direction, and resulting in peeling stress driven failure. This configuration therefore provides conservative failure prediction for composite bonded assemblies compared to other joints. For instance, the reduction in strength for double-lap joints (using ASTM-D3528) would be less than that for the single-lap joints in the presence of the examined bond deficiencies here.

3.1. Bonded assembly and materials

A 2 mm-thickness carbon fibre-reinforced composite laminate was manufactured from aerospace grade unidirectional Hexply® M21/T800S pre-preg using manual laying-up and autoclave curing. The laminate stacking sequence was $[0° \ 90° \ 45° \ -45°]_S$. These were cut to joint lap adherend dimensions (details in Fig. 1 with the dashed line representing the defected bonds).

Joints were bonded as advised by ASTM D5868 [8] using Cytec FM® 94 modified epoxy adhesive film. This aerospace qualified adhesive, which can produce high temperature and good moisture resistant bonds, was applied to the 25 mm × 25 mm overlap region of the adherends. The nominal thickness of the bond was 0.25 mm which is smaller than that in the Standard (0.76 mm). Six categories of single-lap joints with and without bond defects were manufactured, with three specimens per category. These are listed in Table 1 and described below:

1. SB: Standard bonds prepared according to the FM® 94 specification with temperature rate of 2 °C/min.
2. WP: Weak bonds manufactured by pre-curing 20 mm × 20 mm square region of the centre of adhesive area (25 mm × 25 mm) before bonding, on one adherend only. Uncured adhesive added

Table 1
Bond categories in single-lap joints (pressure = 0.28 MPa).

Category	Deficiency method	Label	Cure condition
Standard bond	None	SB	120 °C, 2 °C/min
Weak bond	Bond centre pre-cure	WP	120 °C, 2 °C/min
Weak bond	Rapid heating	WR	120 °C, 4 °C/min
Weak bond	75% reduced cure time	WT	120 °C, 2 °C/min
Kissing bond	Single-side PTFE bond	KS	120 °C, 2 °C/min
Kissing bond	Double-side PTFE bond	KD	120 °C, 2 °C/min

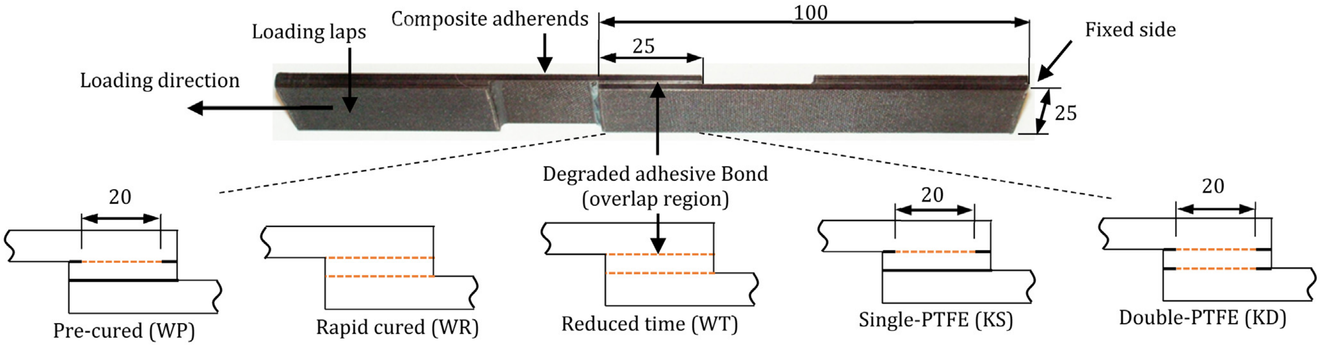

Fig. 1. Single-lap composite bonded joints with defects (all dimensions in mm; bond thickness = 0.25 mm).

to the outer region, 2.5 mm border at perimeter, joints were clamped and then cured.

3. WR: Weak bonds introduced by rapid heating with temperature rate of 4 °C/min.
4. WT: Weak bonds introduced by reducing curing time from 40 min to 10 min (i.e. 75% curing time reduction after reaching 120 °C).
5. KS: Kissing bonds introduced by embedding one 20 mm × 20 mm 0.1 mm-thickness PTFE layer over the centre of the overlap region.
6. KD: Kissing bonds introduced by embedding two 20 mm × 20 mm 0.1 mm-thickness PTFE layers over the centre of the overlap region attached to each adherend.

The PTFE films (categories KS and KD) were embedded at the centre, not at the bond run-outs, to avoid pre-cracking. Single-lap bonded joints experience stress concentrations at the free ends of the overlap region. As the centre of the overlap is approached, the load stress is significantly reduced, i.e. the centre of the bond is ineffective at transferring load [9]. This stress variation along the bond is the reason that the authors decided to affect a large central portion of the bond in WP, KS and KD joints, and enter the high stress region without inserting pre-cracks at free ends (64% of the area was then deteriorated).

3.2. Processing

3.2.1. Surface preparation: peel ply treatment

The surface preparation for all laminates was obtained by use of peel ply. This was removed from the laminate just prior to application of adhesive to provide a contaminant free surface (Fig. 2). The use of peel ply has been commonly used in composite surface preparation as it causes surface roughness, minimises the contamination, and offers a major joint strength in wet conditions, without the risk of damage to the carbon fibres, satisfying both initial and continued airworthiness. The figure shows the peel ply preparation of the composite adherends (Care must be taken during peel ply removal to avoid delamination.).

3.2.2. Adhesive application

A constant pressure of 0.28 MPa was uniformly applied using a rig comprising of mechanical fasteners and clamping plates

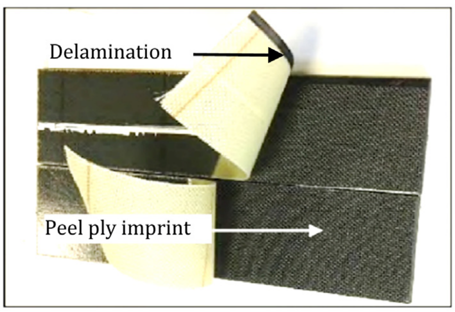

Fig. 2. Peel ply surface preparation of the carbon composite adherends.

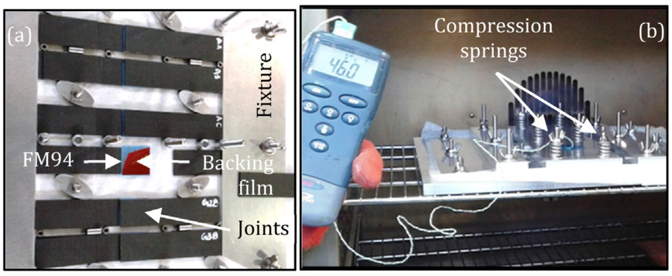

Fig. 3. Process control; (a) pressure fixture, (b) heating monitoring.

(shown in Fig. 3(a)) manufactured to obtain accurate bond lengths and alignment, and uniform bond thickness. The required pressure was then calculated on the basis of the force required to bond six samples by adjusting each compression springs length.

3.2.3. Curing

The SB joints were cured at 120 °C, using a ramp rate of 2 °C/min from ambient, and held at 120 °C ± 5 °C for 40 min in accordance with the adhesive specifications. This was sufficient to reach the bond maximum strength. A thermocouple, attached to the top plate of the jig, ensured temperatures remained constant during curing (Fig. 3(b)).

4. Experiments

The joints were subjected to quasi-static tension performed using a uniaxial test frame fitted with a 30 kN load cell, wedge-action grips and a crosshead speed of 1 mm/min. The joint overlap elongation was measured using a laser extensometer to exclude the compliance of the machine.

5. Results and discussion

5.1. Failure load and displacement

Consistent data were obtained for each bond category, as seen in Fig. 4, for failure load (a) and displacement (b). The KD (double-side PTFE) bond provided the most reproducible data, and WR (rapid cure) and SB provided the largest spread in failure loads especially in displacement data. The results of the WP bond show that despite curing the centre of the bond prior to joining the adherends, the joint was able to sustain 67% of the SB failure load. Rapid curing using the rate of 4 °C/min, twice the specified rate, produced a bond which only reached 50% of the SB failure load. The majority of the load is transferred at the joint ends [9] and as such the pre-cured specimens have near pristine bonding in these regions whereas there is degradation in these areas in the rapidly cured specimens. Among all joints, WT exhibited with the greatest reduction (79%) in the failure load.

All joints were observed to fail at the bond interface (adhesion failure). This was evident from the microscopic images (not shown in the interests of space). The only exception was the WT joint,

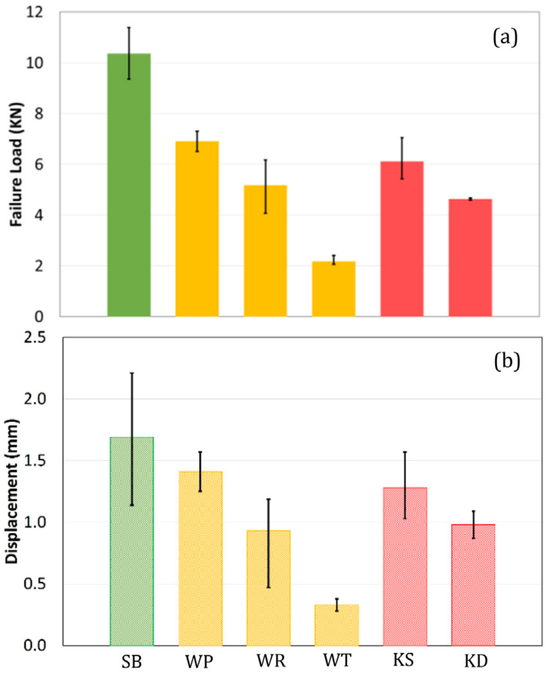

Fig. 4. (a) Failure load, and (b) displacement for all bond categories.

where a combination of interface and cohesive failure was observed. This is expected as in short curing time the adhesive bulk did not reach its ultimate strength.

Note that the adhesive films contain carrier cloth which, based on our observations, did not fail. This is as such believed to be directing damage toward the interface. The bond thickness was relatively low (0.25 mm) compared to the recommendations (0.76 mm). This difference should reduce the joint eccentricity however the reduced thickness bond would be expected to experience higher peel strain. It should be noted that due to the asymmetric bond area in WP and KS, asymmetric secondary bending is introduced, i.e. the composite adherend at the weak bond side undergoes relatively high bending strain.

5.2. ERR-Causality analysis results

Three virtual analyses have been conducted using the ERR-Causality method for WR and WT (curing defects). Note that these are the post-analysis of data, aiming to quantify the effect of curing rate and duration (inputs) on failure load, displacement and energy (outputs). Such parametric correlation analyses will contribute to future curing designs through optimal parameters selection, and applicable to laboratory scale tests.

The first analysis considered failure load as the output. The second one considered failure displacement, and the third considered energy. Considering the quantity of available data, the second order NARMAX model was considered. The candidate model terms are expressed by $\{p_i\} = \{1, u_1, u_2, u_1^2, u_1u_2, u_2^2\}$ where u_1 denotes the curing rate, and u_2 the curing duration. The results are shown in Table 2. As seen, all outcomes are strongly dependent on the curing duration with failure load having the strongest dependence. Moreover the curing rate has shown slight effects on the mechanical response. This does not contradict the findings in Fig. 4 as the model considered the joint performance dependence on the both u_1 and u_2 occurring at the same time. Further investigations are required to ratify though this initially suggests that rapid curing is a better option than reducing duration for cost-effectiveness in joint manufacturing. Also note that this analysis is a proof-of-concept for limited number of parameters and specimens.

Table 2
ERR contribution of each input to the output.

Analysis (outcome, y)	Rate (u_1)	Curing duration (u_2)	Total
1 (failure load)	1.0%	88.2%	89.2%
2 (displacement)	7.7%	68.5%	76.3%
3 (energy)	4.6%	73.5%	78.1%

Table 2 shows that the ERR inputs contribution to three outputs are less than 90%, which suggests that there are other factors that have not been considered. In this sense, the ineffective central region of the bond overlap in WP and KS was also analysed. The model was used to quantify the effect of the degraded area (20 mm × 20 mm). It was found that the bond area reduction by approx. 64% will result in less than 50% failure load reduction (Fig. 4 (a)). Also, the model suggested that there is less than 60% certainty in the reduction, while the certainty of the reduction by curing parameters was >80%. This can be simply attributed to the fact that the high stress gradient in bonded joints occurs at the bond run-outs, not at the centre, and then stresses the importance of processing parameter control in determination of the mechanical performance of final assembly.

6. Conclusions

The current research provided a comparative study of adhesively bonded joints with bond deterioration in order to address the importance of process parameters in aerospace composite bonding procedures. A nonlinear correlation analysis, ERR, was used to quantify the effect of curing parameters on the joints failure. A number of techniques were used to introduce defects to the bond area of the single-lap bonded joints. It was found that a joint with any defect, non-standard curing or contaminant, has a reduced strength. The 'kissing bond' defect was found to cause the lowest 'failure load reduction' (categories WP and KS with failure load reduction of 33% and 41%). Curing time had the most significant effect on the bond response. This, based on the ERR analysis, was also the strongest process-linked parameter by >70% dependency level of joint response (Table 2). This may become extremely important for relatively large composite bonded assemblies with unsymmetrical geometric features (e.g. in aircrafts) where non-uniform heating is present. The study showed that control of the curing process is as important as ensuring good surface preparation when producing adhesively bonded assemblies.

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