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A design of experiments assessment of moisture content in uncured adhesive on static strength of adhesive-bonded galvanized SAE1006 steel

Jianping Lin[a,*], Zhiguo Lu[a], He Yang[a], Peichung Wang[b]

[a] School of Mechanical Engineering, Tongji University, N4800, Caoan Highway, Shanghai 201804, China

[b] Research & Development Center, General Motors Corporation, Warren, MI 48090-9055, USA

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ABSTRACT

As part of a cooperative research program to develop and implement crash-resistant toughened adhesives targeted for future vehicles, this paper summarizes a study of the influence of pre-exposure of uncured adhesive and steel sheets in a humid and elevated temperature environment on quasi-static strength of bonded hot dipped galvanized SAE1006 steel joints.

In this study, we use a DOE (design-of-experiment) program called DEXPERT to design the experiment and to analyze the effects of exposure temperature, exposure time, curing temperature and curing time on joint strength of adhesive-bonded galvanized SAE1006 steel. Prior to adhesive curing, the adhesive and galvanized steel coupons were pre-exposed to various relative humidity levels and temperatures. The experimental results were then analyzed by DEXPERT and the relative contributions of each factor on variance in joint strength were calculated. It was found that curing temperature is the most influential factor affecting the strength of adhesive-bonded galvanized SAE1006 steel joints. The curing of a joint at 180 °C can increase the robustness of the process and provides the greatest strength regardless of the variation of other factors. The joint strength curing at 150 °C shows a strong sensitivity to the curing time, while the adhesive cannot cure at 130 °C at all under all conditions. It has also been found that the pre-exposure of adhesive and steel for an hour can slightly decrease the joint strength at high temperature and humidity. Therefore, the effect of long time exposure of the uncured adhesive and steel still needs to be further investigated.

1. Introduction

Epoxy based adhesive-bonded steel has a wide range of applications in light-weight vehicle structures for good stiffness and strength [1,2]. To obtain a good bonding strength, the bonded steel components in the vehicles must be held at a certain temperature for a certain length of time to reach full cure [3–6]. This curing process is often performed in the paint shop without a dedicated curing oven for cost consideration. In the paint shop, the ambient curing temperature and the curing time are two main variables, which can significantly influence the adhesive cure and the bonding strength [7,8]. Before the curing of the bonded steels, the adhesive and steels could be exposed to the ambient hot humid environment due to the variable weather conditions, since the bonded components cannot immediately come into paint shop after adhesive was dispensed on the steels. It is known that the humid environment can lead to the strength degradation of adhesive bonded steels [9–11]. In the past, most studies have been undertaken on the effects of environmental condition on the mechanical properties of cured adhesive-bonded steels [12–14]. These researches provide valuable information on the humid environmental aging of adhesive bonded steel. However, little information is available concerning the effects of exposure of uncured crash-resistant toughened adhesive in hot humid environment.

In this study, the combined effects of pre-exposure of uncured adhesive and steel in hot humid environment and the curing conditions on the quasi-static strength of cured crash-resistant toughened adhesive-bonded steels are investigated. To understand the combined effects of pre-exposure and the curing, we use a DOE (design-of-experiment) program called DEXPERT [15] to design the experiment and to analyze the effects of exposure temperature, exposure time, curing temperature, curing time and humidity levels on joint strength of adhesive-bonded galvanized SAE1006 steel. Adhesive is first dispensed on a steel sheet and both dispensed adhesive and steel adherends are exposed in 65% or 95% relative humidity at 20 or 40 °C for a period of 0, 10 or 60 min. After curing, mechanical tests were conducted in ambient environment. In this study we used lap-shear specimens made from 0.75 mm thick galvanized SAE1006 steel and adhesive. The details of the experimental design, sample fabrication and data analysis are presented in the following section of this study.

* Corresponding author. Tel.: +86 139 0171 9457; fax: +86 021 69589485.
E-mail address: jplin58@tongji.edu.cn (J. Lin).

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This study is a part of a larger program to develop a more quantitative and predictable description of environmental durability of crash-resistant toughened adhesive-bonded steels.

2. Design of experiment

For high volume automotive applications, simpler, faster and more environmental friendly processes are required. Normally, the sheet metal is covered with oil during transportation from steel mill to stamping plant. After delivery to the plant the steel is sheared and stamped. The next stage of the process is the application of the adhesives for weld-bonding. This is achieved by using equipment of varying complexity, from hand-operated pumps to sophisticated robotic dispensing systems. The robot is programmed to apply a bead of adhesive. After the adhesive has been applied, the panels are married together to form the parts. The panels are then put through paint oven with temperature ranging from 180 to 200 °C. The typical time in the oven is from 20 to 30 min. Since there are many processing parameters (e.g., temperature and humidity in the plant, curing temperature and curing time in paint oven) to control in this production environment, it is conceivable that bonded components may experience a range of humidity level, curing temperature and time. Therefore, it is essential that an understanding of the effects of these variables on the strengths of bonded joints be obtained. The present study was undertaken to experimentally evaluate the effects of these process variables.

Table 1 shows the input factors and responses for this experiment. The design consists of five input factors—exposure time, exposure temperature, curing time, curing temperature and humidity level. There is only one response variable, namely joint strength. Shown in Table 1 are the details of the input factors as defined in DEXPERT. Curing temperature, curing time and exposure temperature each have 3 levels. Exposure time and humidity have only two levels. This DOE is a full factorial design with 3 replicates, which requires a total of 324 specimens.

3. Experimental procedure

3.1. Material

Low carbon hot-dipped galvanized (HDG60) steel SAE1006 was used in this study. Chemical composition and mechanical properties of the steels are shown in Table 2 and Fig. 1(a), respectively. The adhesive used in this study was Henkel one-part adhesive, a proprietary crash-resistant toughened epoxy. Bulk adhesive specimens were fabricated (based on manufacturer's recommended curing procedure (i.e., 180 °C and 30 min)) and tested. Fig. 1(b) shows the mechanical properties of the adhesive.

Table 1
Details of input factors in DEXPERT.

Factor	Level
Curing time (min)	10, 15, 20
Curing temperature (°C)	135, 150, 180
Exposure time (min)	0, 10, 60
Exposure temperature (°C)	20, 40
Humidity (%)	65, 95

3.2. The environmental chamber

To simulate the extended exposure in the humid environment, the adhesive was dispensed on a steel adherend and then exposed in a laboratory environmental chamber (0.6 m × 0.6 m × 1.5 m) as shown in Fig. 2. Dispensed adhesive and steel adherends were suspended in the humidity chamber. The water vapor was supplied through the humidifier. The vapor is added in the form of steam by boiling faucet water in a stainless steel tank maintained at a constant temperature. The vapor is pumped into the chamber and is circulated inside the chamber by means of a circulating fan (placed inside the chamber) to maintain a uniform environment throughout the chamber. The relative humidity (R.H.) inside the chamber is controlled by means of an optical dew point hygrometer. Selected workpieces were periodically removed from the humidity chamber for joint fabrication.

3.3. Specimen fabrication

The lap-shear specimen configuration, shown in Fig. 3, was fabricated from 38 mm × 127 mm hot-dipped galvanized steel sheets (SAE1006). To simulate the production environments, the steel sheets used in this study were not specifically cleaned (i.e., as-received condition). Shims are bonded on the adherends to keep the load plane of the specimen coincident with central plane of the tensile tester. The adhesive-bonded specimens were prepared as follows: (1) applying the adhesive through a hand-held injection gun on one of the two adherends, which were stored in an ambient laboratory environment (20 °C and 50% R.H.); (2) positioning the adherends with and without dispensed adhesive with a fixture in the humidity chamber; (3) after removing the steel adherends from the humidity chamber the adherends were brought together by a fixture under ambient laboratory conditions; (4) applying the pressure through the fixture so that a adhesive thickness (set up by a 0.25 mm thick metal shim) of 0.25 mm can be maintained; (5) curing the specimens in the oven for period of a time and temperature as prescribed by to DOE schedule. All finished specimens are examined and the spew fillets around the edge of the overlap were remained to simulate the real production conditions.

3.4. Static testing

Static tests were performed by loading each specimen to failure in a tensile tester. To minimize bending stresses inherent in the testing of lap shear specimens, filler plates were attached to both ends of the sample using masking tape to accommodate the sample offset. Load vs. displacement curves were obtained as the specimens were loaded at a stroke rate of 2 mm/min. Three replicates were tested, and the average peak loads were reported.

Table 2
Chemistry (wt%), coating and sheet gage for SAE1006 steel.

Steel	C	Mn	P	Si	Ni	S	Al	Cr	Ca	Ti	Gage (mm)
SAE1006 (HDG60)	0.006 max.	0.2 max.	0.025 max.	–		0.02 max.	0.015 min.	–	–	0.03/0.08	0.75

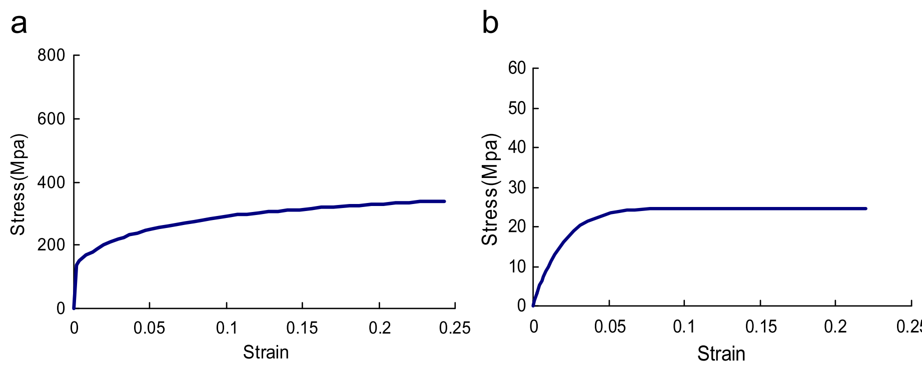

Fig. 1. Stress-strain properties of (a) SAE1006 steel and (b) Henkel one-part adhesive.

Fig. 2. Photographs of the environmental chamber.

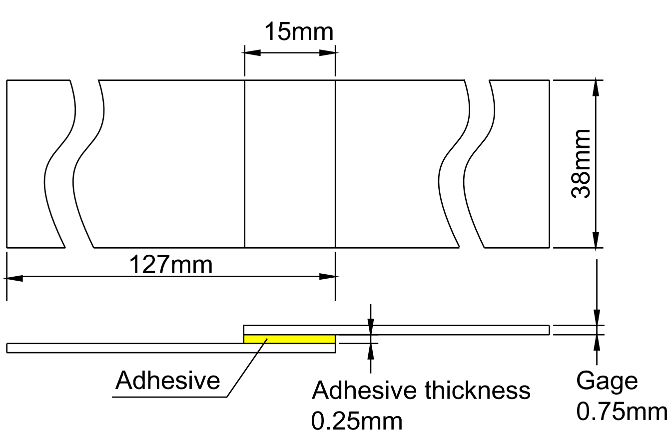

Fig. 3. Schematic of a lap-shear joint.

4. Results and discussion

The analytical capability in DEXPERT [16] includes the analysis of variance, variance component estimation, percent contribution, comparison of means and polynomial approximation. Analysis results are displayed in either a tabular or graphical format.

4.1. Main effect factors and interactions

The main effects of the input factors and their interactions on the response variable are shown in Table 3 and Fig. 4 for the response variable-joint strength. The main effect of a particular factor is calculated over the entire range of levels for all other factors. This means that the indicated effect for a given factor is the average response obtained for this factor when averaged over all combinations of settings of all other input factors. Shown in Table 3 is the analysis of variance for all main factors and their interactions. Column 3 – *P*-value – is the probability value for each term. *P*-value indicates the probability that the effect caused by a given term is due merely to random chance. The smaller the *P*-value, the less likely the effect is random and the more significant the term is. A double asterisk (**) in the *P*-signif. column indicates the term is extremely significant, a single asterisk (*) indicates a very significant term and the absence of an asterisk indicate the term is of little significance. The single factors and interactions with little significance are not listed in Table 3.

These ANOVA results are displayed graphically in Fig. 4. As shown in Fig. 4(a), as the curing temperature increases, there is an increase in average joint strength. This increase is particularly pronounced for curing temperature between 150 and 180 °C.

Table 3
Analysis of variance summary of joint strength for bonded galvanized SAE1006 steel.

Term	*P* value	*P* signif.
Curing temperature	1.1119E−177	**
Curing time	1.9478E−54	**
Curing temperature × curing time	8.9655E−67	**
Curing temperature × curing time × exposure time × humidity	0.030373	*
Exposure temperature × exposure time	0.035057	*
Error	…	

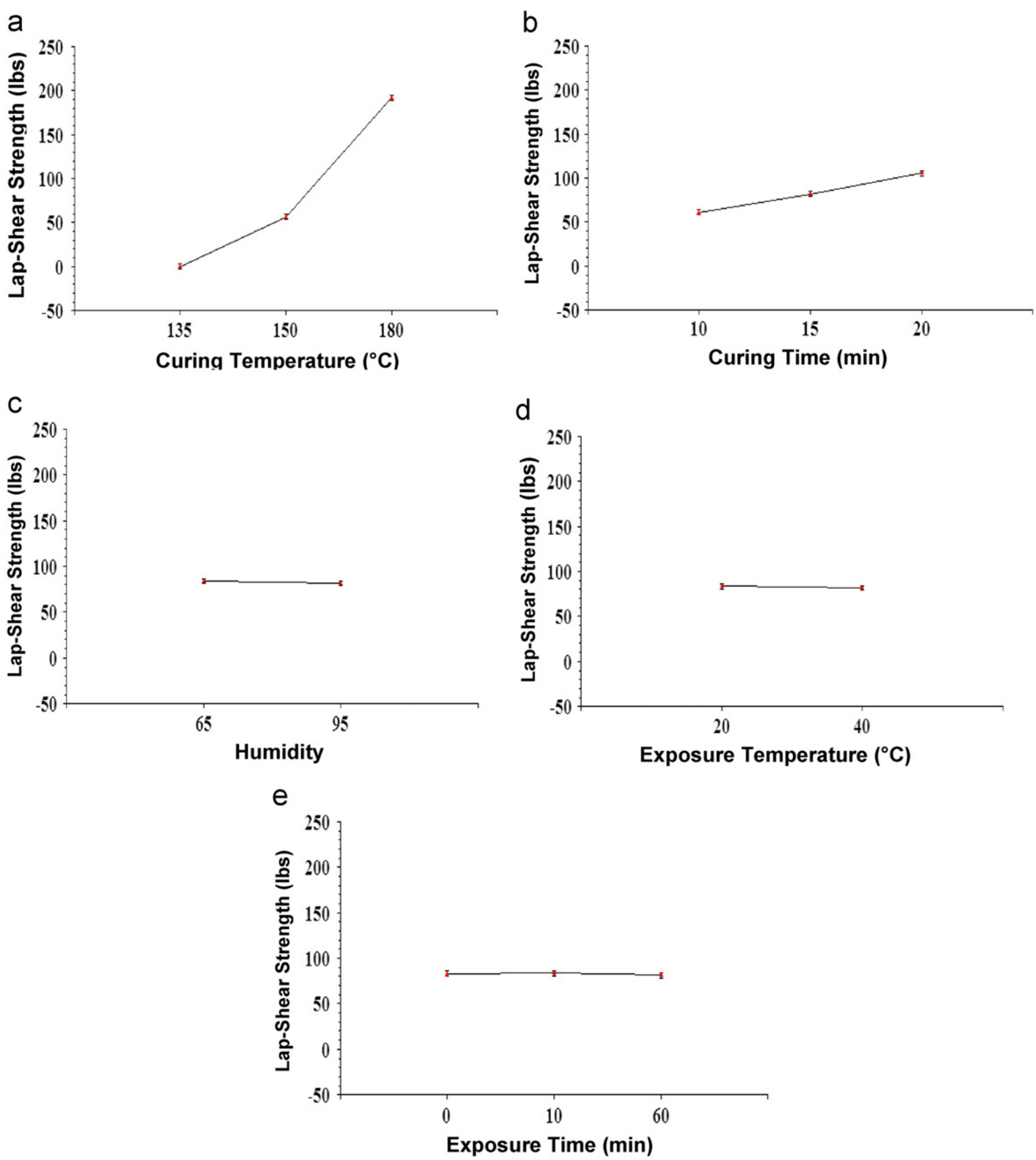

Fig. 4. Effect of (a) curing temperature, (b) curing time, (c) humidity level, (d) exposure temperature and (e) exposure time on joint strength of bonded galvanized SAE1006 steel.

The effect of curing time on average joint strength is shown in Fig. 4(b). The average joint strength increases with an increase in curing time. Average joint strength increases approximately in a linear fashion with increasing curing time between 10 and 20 min.

The effect of humidity level on average joint strength is shown in Fig. 4(c). As shown, by increasing the humidity level from 65% to 95%, average joint strength decreased slightly. Similar results shown in Fig. 4(d) and (e) were observed for exposure temperature and exposure time, respectively.

Table 4
% contribution of factors and interactions on total variance of joint strength for adhesive-bonded galvanized SAE1006 steel.

Term	Variance components	% Contribution
Curing temperature	9692.9	83.2
Curing temperature × curing time	1109	9.52
Curing time	486.73	4.18
EMS(error)	228.13	1.96
Curing temperature × curing time × exposure time × Humid	44.727	0.384
Curing temperature × curing time × exposure time × exposure temperature	36.074	0.31
Exposure temperature × exposure time	10.153	0.0872
Curing time × exposure time × humidity	10.065	0.0864
Curing temperature × exposure temperature × exposure time	9.8078	0.0842
Exposure temperature × exposure time × humidity	6.707	0.0576
Curing temperature × exposure temperature × exposure time × humidity	5.7029	0.049
Humidity	2.8249	0.0243
Curing temperature × exposure temperature × exposure time	2.764	0.0237
Curing temperature × exposure temperature	0.1217	0.00104

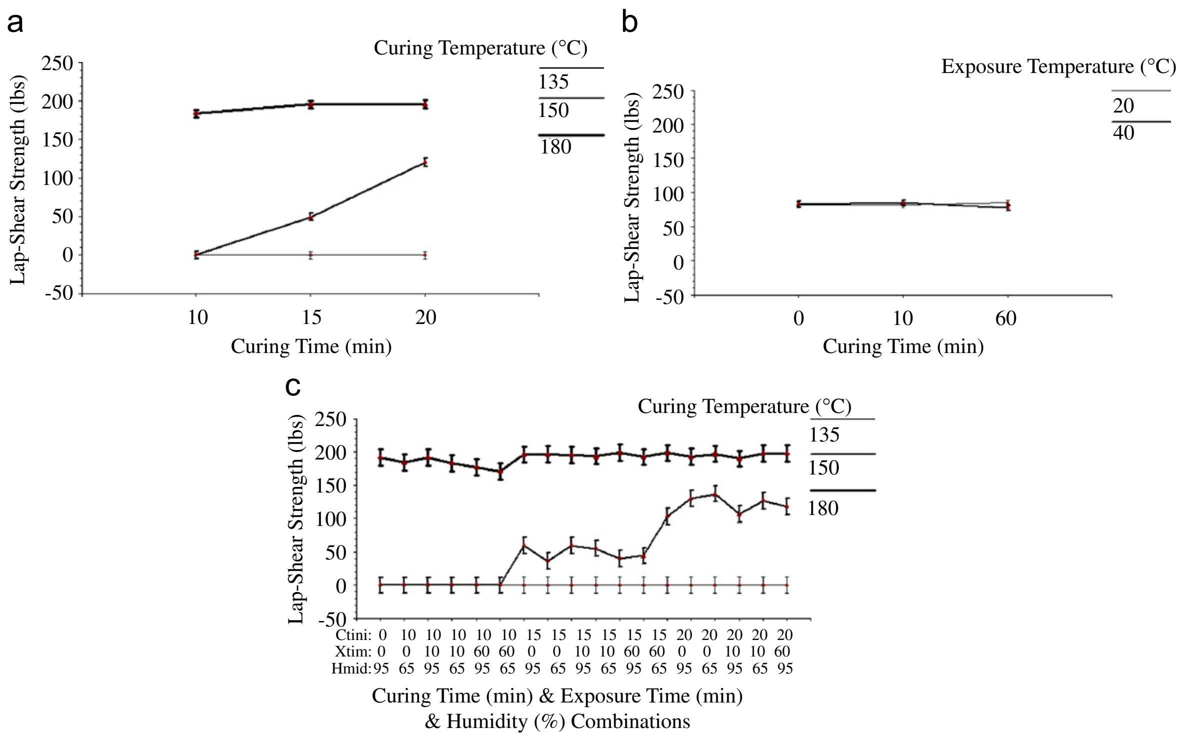

Fig. 5. Effect of the interaction between factors on the strength of bonded 0.75 mm thick galvanized SAE1006 steel: (a) curing temperature and time, (b) exposure temperature and time and (c) curing temperature, curing time, exposure time and humidity level.

4.2. Analysis of variance

The basic idea of ANOVA is to explain the variation in the response variables. The variation can be due to the main effects and/or interaction effects, with the random error term accounting for the rest. This can be used to determine whether or not each term has a significant effect on the response variable. Table 4 shows the basic ANOVA results for the data collected in this experiment (single factors and interactions with no contribution are not listed).

As shown in Table 4, curing temperature is the most influential input factor. Although curing time and the interaction of curing temperature and time contribute to the variation in average joint strength, their influences are much smaller than the effect of curing temperature. These results are of particular importance due to the manufacturing process in which the bonded steel is applied. To obtain the optimum adhesive bond strength, the curing temperature needs to be properly controlled.

4.3. Interaction between factors

As shown in Table 4, the curing temperature and the interaction of curing temperature and time were identified as the biggest contributors to variation in joint strength. Curing temperature contributed 83.2% and interaction of curing temperature and time contributed 9.2% to total joint strength variation. Unknown factors (EMS[error]) accounted for 1.96% variation. The effects of significant terms are shown graphically in Fig. 5. Fig. 5(a) shows the effect of the interaction of curing temperature and curing time on joint strength. As shown, for a curing temperature of 180 °C, changes in curing time in the range of 10–20 min had little influence on joint strength variation. This curing temperature also resulted in highest joint strength. This indicates that curing the adhesive at 180 °C increases the robustness of the process and provides the greatest strength. However, for the curing temperature of 150 °C, as the curing time increases, there is a somewhat linear increase in average joint strength. This indicates a strong sensitivity to curing time. Curing the adhesive at 150 °C results in a less robust process. For the curing time of 135 °C, the joint strength is almost zero for all curing times. This result indicates the adhesive cannot cure at 135 °C at all.

The individual main effects of the exposure temperature and exposure time on average joint strength were shown in Fig. 4(d) and (e), respectively. When viewing these figures, it is not apparent that an interaction exists between these two factors. However, by performing a DOE analysis of the data the interaction is revealed and shown in Fig. 5(b). When the exposure temperature is 20 °C, joint strength increases slightly as exposure time increases from 0 to 60 min but, when the exposure temperature is 40 °C there is a slight decrease in joint strength with increasing exposure time. This result shows that a short exposure in less than an hour can slightly decrease the strength of adhesive bonded steel at a relatively high exposure temperature. Therefore, the effect of long time exposure on the uncured adhesive and steel still needs to be further investigated.

Fig. 5(c) shows the 4-way interaction between the curing temperature, curing time, exposure time and humidity level. When the curing temperature is 135 °C joint strength is 0 under all conditions indicating the adhesive did not cure. When the curing temperature is at 150 °C and curing time is 10 min joint strength is again 0 regardless of the level of exposure time and humidity. When curing time is increased to 15 min, joint strength increases to an intermediate level. This level is only slightly affected by changes in exposure time and humidity. Joint strength increases again when the curing time is increased to 20 mins Again joint strength is only slightly influenced by changes in exposure time and humidity. Finally, when the curing temperature is set to 180 °C joint strength reaches to a maximum. At this temperature joint strength is relatively unaffected by the levels of the other factors. This demonstrates that the process is most robust at a curing temperature of 180 °C.

4.4. Full DOE regressional model

To show the effects of input factors on average joint strength, a full DOE regressional was generated. The effects of various input factors are reproduced by a regressional analysis and the results

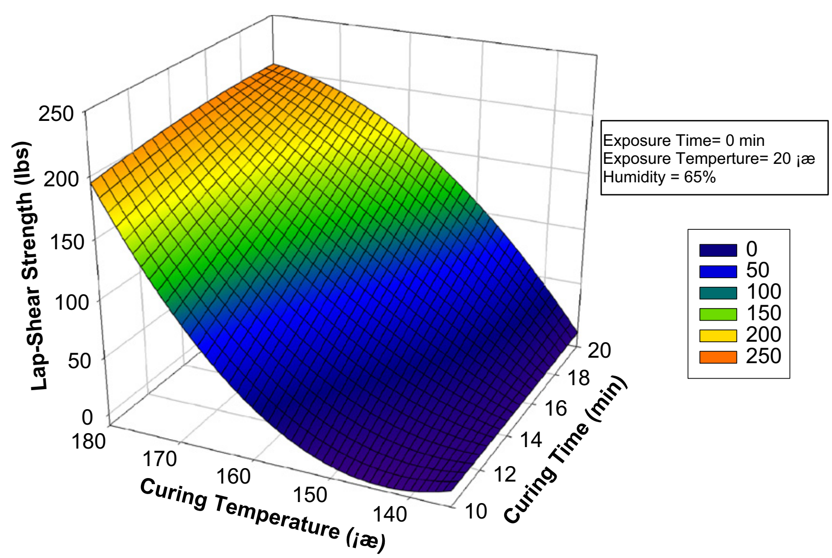

Fig. 6. Effect of curing temperature and time under ambient conditions on the strength of bonded 0.75 mm thick galvanized SAE1006 steel joints.

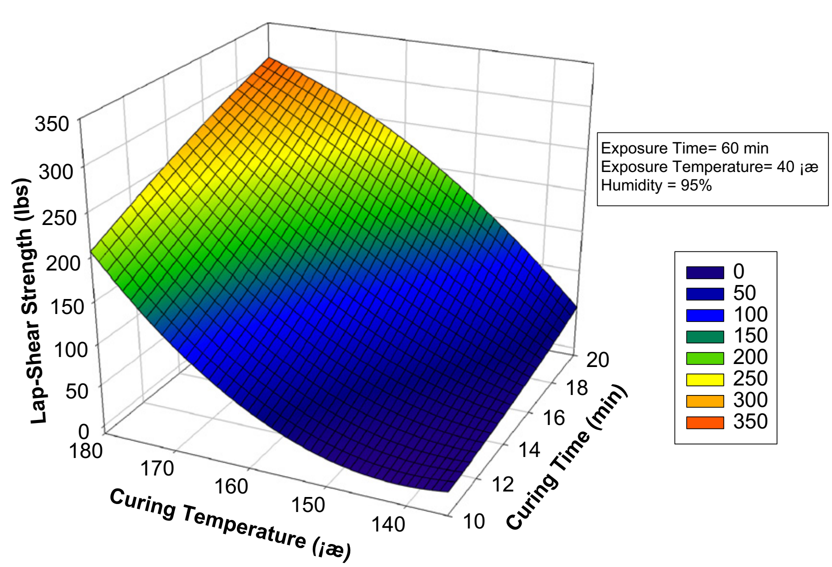

Fig. 7. Effect of combined curing temperature, curing time, exposure temperature, time and humidity level on the strength of bonded 0.75 mm thick galvanized SAE1006 steel joints.

are shown in Figs. 6, 7, 8 and 9. It is seen that within the selected range of input factors, curing temperature is the most influential factor on joint strength. The effect of curing time is smaller than that of the interaction of curing temperature and curing time, but greater than exposure temperature and time.

The results presented in this study illustrate the effects of humidity level, exposure temperature, exposure time, curing temperature and curing time on the joint strength of bonded galvanized SAE1006 steel. While the present results show that reduction of joint strength due to the exposure before the curing is negligible during assembly process, care should be taken in proper control of curing temperature and curing time. Since the temperature distribution in the curing oven is usually uneven, the bonded structures at different parts of vehicle body can encounter very different curing temperatures. It is strongly recommended the process development and plan used in vehicle assembly should be considered and adjusted to account for possible adverse effects due to uneven temperature distribution in the oven. In addition, note that it is very probable a great strength decrease would occur if both the adhesive and steel were pre-exposed to a hot-humid environment for a longer time. Further investigation is still needed for providing detailed information concerning the effect of pre-exposure before the curing on the strength of adhesive bonded steel.

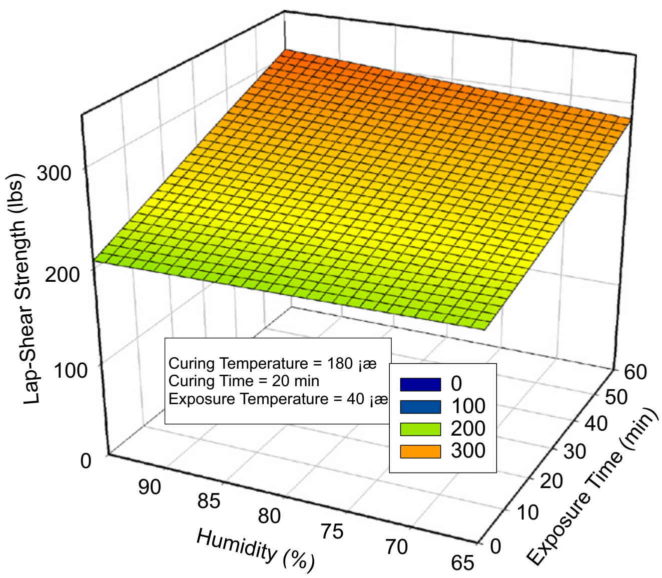

Fig. 8. Effects of exposure time and humidity level at 40 °C on the strength of bonded. 75 mm thick galvanized SAE1006 steel joints.

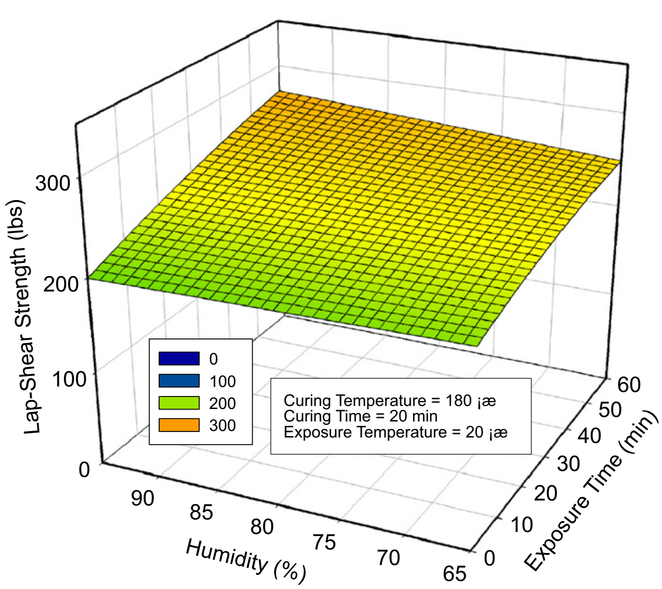

Fig. 9. Effects of exposure time and humidity level at ambient temperature on the strength of bonded 0.75 mm thick galvanized SAE1006 steel joints.

5. Conclusions

To understand the combined effects of pre-exposure and the curing conditions, a DOE (design-of-experiment) conducted to analyze the effects of humidity level, exposure temperature, exposure time, curing temperature and curing time on joint strength of adhesive-bonded 0.75 mm thick galvanized SAE1006 steel with crash-resistant toughened adhesive concluded the following:

1. With the variables and range studied here, the curing temperature had the greatest effect on joint strength of adhesive-bonded 0.75 mm thick galvanized SAE1006 steel. The effect of curing time is not as significant as that of the curing temperature but is greater than that of exposure temperature and exposure time.
2. The joint strength increases with curing temperature over a range of 135–180 °C. This increase is particularly pronounced for curing temperature between 150 and 180 °C. When the curing temperature is 135 °C, joint strength is always 0 under all conditions, showing the adhesive cannot cure at this temperature.
3. The effect of curing time on the joint strength is dependent on the curing temperature. The joint strength shows a strong sensitivity to curing time when curing temperature is 150 °C. When curing the joint at 135 and 180 °C, the effect of curing time is not so significant.
4. A short exposure for less than an hour can slightly decrease the strength of adhesive bonded steel at a relatively high temperature. Therefore, the effect of pre-exposure when exposure time is longer still needs further investigation.

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