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Thermal stress-induced thermoplastic composite debonding, studied by contact electrical resistance measurement

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Abstract

Thermal stress-induced debonding of polyphenylenesulfide from polyphenylenesulfide was studied nondestructively by measurement of the contact electrical resistance between members containing continuous unidirectional carbon fibers at 90°. The resistance increased by up to 600% upon debonding. The resistance increase was much greater than the resistance decrease during prior bonding. Debonding occurred during cooling when the pressure or temperature during prior bonding was not sufficiently high. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

Engineering thermoplastics can be bonded together by solid state welding (i.e., interdiffusion or autohesion above the glass transition temperature but below the melting temperature) or fusion welding (i.e., melting and subsequent solidification). Both methods involve heating and subsequent cooling. During cooling, the thermoplastic goes from a soft solid state (in the case of solid state welding) or a liquid state (in the case of fusion welding) to a stiff state. If the thermoplastic members to be joined are anisotropic (as in the case of each member being reinforced with fibers) and the fiber orientation in the two members is not the same, the thermal expansion (actually contraction) mismatch at the bonding plane will cause thermal stress to build up during cooling. This thermal stress is detrimental to the quality of the adhesive bond formed between the two members.

Although the detrimental effect of thermal stress to a joint is well-known, actual debonding induced by thermal stress has not been previously investigated. Two scenarios can lead to the absence of bonding after cooling. One scenario is the absence of bond formation at the high temperature during welding, due to insufficient time or temperature. The other scenario is the presence of bonding at the high temperature, but the occurrence of debonding during subsequent cooling due to thermal stress. The cause of the absence of bonding is different in the two scenarios. In any given situation, the cause of the debonded joint must be understood if the absence of bonding after cooling is to be avoided.

The propensity for mutual diffusion in thermoplastic polymers increases with temperature. The contact at the interface across which interdiffusion takes place also plays a role. An intimate interface, as obtained by application of pressure to compress the two members together, also facilitates diffusion. Thus, the quality of the joint improves with increasing temperature and increasing pressure in the high-temperature period of welding. The poorer is the quality of the joint attained at high temperature, the greater is the likelihood that thermal stress built up during subsequent cooling will be sufficient to cause debonding. Hence, merely having bonding achieved at the high temperature in welding is not enough. The bond achieved must be of sufficient quality to withstand the abuse of thermal stress during subsequent cooling.

The quality of a joint is conventionally tested destructively by mechanical methods or nondestructively by ultrasonic methods [1, 2]. This testing is performed at room temperature after the joint has been cooled from the high-temperature used in welding. As a result, the testing does not allow distinction between the two

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scenarios described above. This paper describes the use of a nondestructive method, namely contact electrical resistance measurement, to monitor joint quality in real time during the high-temperature period of welding and also during subsequent cooling.

The electrical nature of this method requires the presence of an electrically conductive filler, such as carbon fiber, in each thermoplastic member to be joined, since the thermoplastic itself is not conductive. The higher is the quality of the joint, the greater will be the number of contacts between the filler on the two sides of the joint interface, and the lower will be the contact electrical resistance of the interface. This method has previously been used to monitor solid state welding in thermoplastics during the high-temperature bonding period [3]. Such monitoring allowed a study of the kinetics of the bonding process as the contact electrical resistivity continuously decreased during bonding. This paper extends the work from the high-temperature bonding period to the subsequent cooling period, in which debonding due to thermal stress may or may not occur. Debonding is indicated by a significant increase in the contact electrical resistance.

This paper also correlates the temperature and pressure in the high-temperature bonding period with the occurrence of debonding during subsequent cooling. This correlation provides guidelines for the choice of temperature and pressure for thermoplastic welding in industrial practice, even though the exact temperature and pressure that are appropriate vary with the composition and anisotropy of the thermoplastic-matrix composite (as the composition and anisotropy affect the thermal stress).

This paper also confirms the value of contact electrical resistance measurement as a nondestructive in situ method for testing the bonding of thermoplastics. The method is relevant to quality control in manufacturing that involves bonding of thermoplastics. Furthermore, the electrical output of this method makes the method amenable to the closed-loop form of quality control. In other words, during manufacturing, the temperature and pressure used for bonding may be adjusted in real time in response to the result of the electrical resistance measurement.

Because of their high strength-to-weight and stiffness-to-weight ratios, carbon fiber-polymer matrix composites are effective materials for lightweight structures such as aircraft, automobiles, sporting goods and wheel chairs. The thermoplastic-matrix composite members to be joined in this work are composed of continuous unidirectional carbon fibers in a polyphenylenesulfide (PPS) matrix. The electrical conductivity of the carbon fibers makes the electrical method feasible. Each member is a single layer (lamina). The fibers in the two members are at an angle of 90° to each other. The interface where bonding takes place is the interface between the two

crossed laminae. The unidirectionality of the fiber makes each lamina strongly anisotropic, which results in thermal stress at the interface during cooling from the high-temperature of welding.

It should be noted that the contact electrical resistance of the interface is in the through-thickness direction. In spite of the insulating nature of the polymer matrix, this resistance is always finite when bonding occurs at the interface. This is because the contact between individual fibers of the two laminae develops whenever bonding occurs. This contact is made possible by small scale relative motion of fiber and polymer molecules during the evolution of bonding. Development of the adhesive bond increases the extent of the fiber–fiber contact, thereby decreasing the contact electrical resistance. Debonding decreases the extent of fiber–fiber contact, thereby increasing the contact resistance. This notion is similar to that behind the use of through-thickness volume electrical resistance measurement to monitor delamination in a continuous carbon fiber epoxy-matrix composite [4]. Contact resistance rather than volume resistance is studied in this work, because the contact resistance directly relates to the quality of the interface (bond) whereas the volume resistance reflects the resistance of each lamina as well as the interface between laminae.

In this work, the temperature and pressure (in the through-thickness direction) of the composite members at 90° during bonding in the high-temperature stage of welding were varied. For each experiment, the quality of the bond was monitored by contact electrical resistance measurement during (a) bonding at a constant temperature (above the glass transition temperature) and pressure and (b) subsequent cooling. Confirmation of bonding and debonding was also obtained by destructive mechanical testing involving manual peeling performed at the high-temperature prior to cooling, as well as after cooling. Bonding attained at all temperatures and pressures used for the high-temperature stage. However, debonding upon subsequent cooling occurred when either the temperature or the pressure was not high enough in the high-temperature stage.

2. Experimental methods

The thermoplastic used was polyphenylenesulfide (PPS). Its glass transition temperature (T_g) was 90°; its melting temperature (T_m) was 280°C. The material was in the form of continuous unidirectional carbon fiber prepreg supplied by Quadrax Corp. (Portsmouth, Rhode Island; Product QLC4164). The thickness of the prepreg was 250 μm. the carbon fiber within the prepreg was AS-4C, from Hercules Advanced Materials and Systems Company (Magna, Utah), with diameter 8 μm. The fiber weight fraction was 64%.

The prepreg was used after annealing. Annealing was carried out in air at 180°C for 25 h, while pressure (1000 Pa) was applied by the weight of steel plates. After annealing, the prepreg was furnace-cooled to room temperature under pressure.

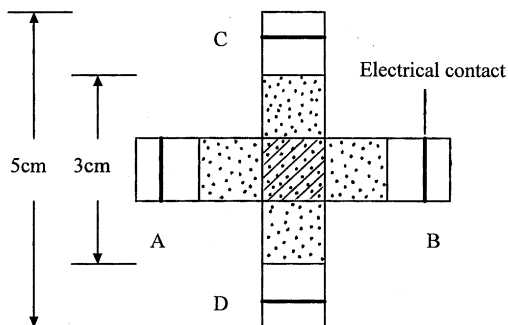Prepreg strips 5 cm in length and 1 cm wide were placed on one another at an angle of 90° in a cross-shaped steel mold cavity lined with a PTFE film for electrical insulation, The overlap area was 1 cm × 1 cm, as shown in Fig. 1. A pressure was applied through a 3-cm long cross-shaped steel plate, which was electrically insulated from the prepreg by a PTFE film. An electrical contact in the form of silver paint in conjunction with copper wire was applied at each of the four legs of the crossed prepreg strips (Fig. 1). Two of the electrical contacts (A and D in Fig. 1) were for passing current; the remaining two contacts (B and C) were for measuring voltage. The current flowed from current probe A along one lamina, turned to the through-thickness direction and flowed through the junction from one lamina to the other, and then turned direction again to flow along the other lamina toward current probe D. The voltage between probes B and C gave the voltage across the junction. The potential at B was higher than that of C since the current flowed from A to D. The voltage divided by the current gave the contact resistance of the joint. This constitutes the four-probe method of DC electrical resistance measurement. A Keithley 2001 multimeter was used.

Samples as illustrated in Fig. 1 were heated from 30°C to different temperatures (in the range from 240 to 290°C)

Fig. 1. A sample configuration for studying the kinetics of bonding. The dotted cross-shaped region is where pressure is applied. The square-shaded region is the joint.
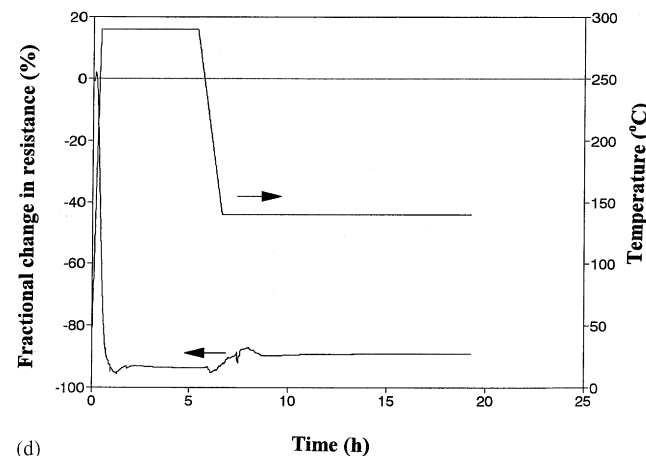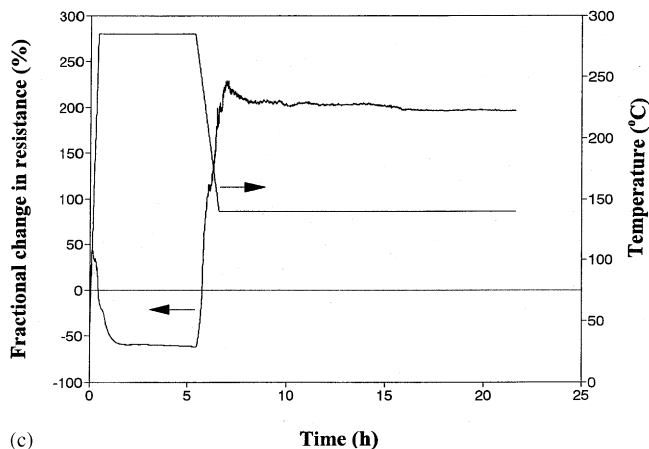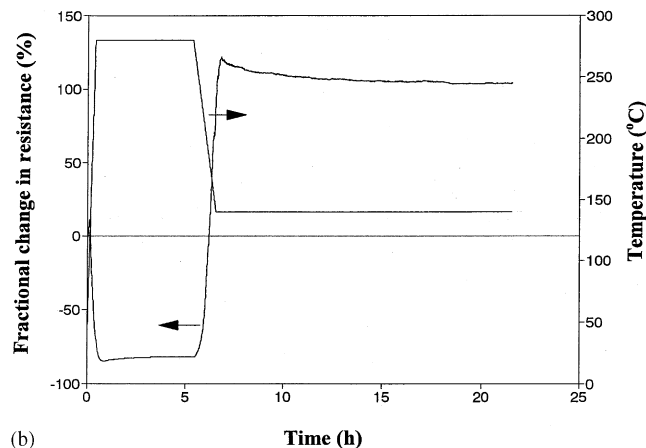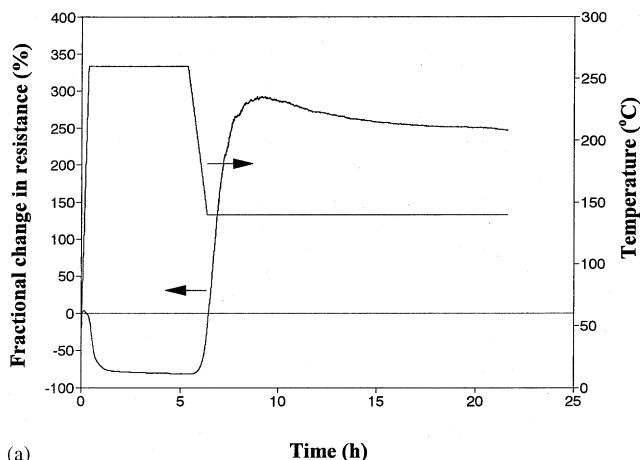

Fig. 2. The effect of bonding temperature on the contact resistance variation at a fixed pressure of 4.8×10^3 Pa. (a) 260°C. (b) 280°C. (c) 285°C. (d) 290°C.

at a heating rate of 10°/min and then were held at the temperature for either 5 or 15 h in order to allow bonding to occur. After that, the samples were cooled to 140°C at a rate of 2°C/min and then were held at 140°C for 15 h. During the temperature variation, the contact resistance was continuously measured.

3. Results and discussion

Fig. 2 shows the effect of bonding temperature (i.e., temperature in the high-temperature stage) at a fixed pressure of 4.8×10^3 Pa. For all bonding temperatures other than 290°C (the highest temperature), debonding occurred upon subsequent cooling. The debonding was accompanied by a sharp rise in the contact resistance, as shown in Fig. 2(a)–(d). The absence of debonding yielded a negligibly small rise in the contact resistance, as shown in Fig. 2(e). Hence, at a pressure of 4.8×10^3 Pa, the minimum bonding temperature for avoiding debonding during subsequent cooling was 290°C — slightly above the melting temperature of PPS.

After debonding, the resistance was even higher than the value measured for the joint prior to bonding. This behavior is attributed to the formation of cracks which degraded the electrical contact, thus causing the resistance to increase dramatically.

Fig. 3 shows the effect of bonding pressure (i.e., pressure in the high-temperature stage) at a fixed bonding temperature of 270°C. For the lowest pressure of 4.8×10^3 Pa, debonding occurred upon cooling subsequent to bonding. Debonding was accompanied by a sharp rise in the contact resistance, as shown in Fig. 3(a). The absence of debonding yielded a negligibly small rise in the contact resistance, as shown in Fig. 3(b) and (c) for the bonding pressures of 3.0×10^5 and 6.8×10^5 Pa. Hence, at a bonding temperatur of 270°C, the minimum bonding pressure for avoiding debonding during subsequent cooling was found to be 3.0×10^5 Pa.

The increase in resistance during debonding was much greater than the decrease in resistance during bonding, as shown in Figs. 2 and 3. The fractional increase in resistance during debonding was a high as 600% (Fig. 3(a)).
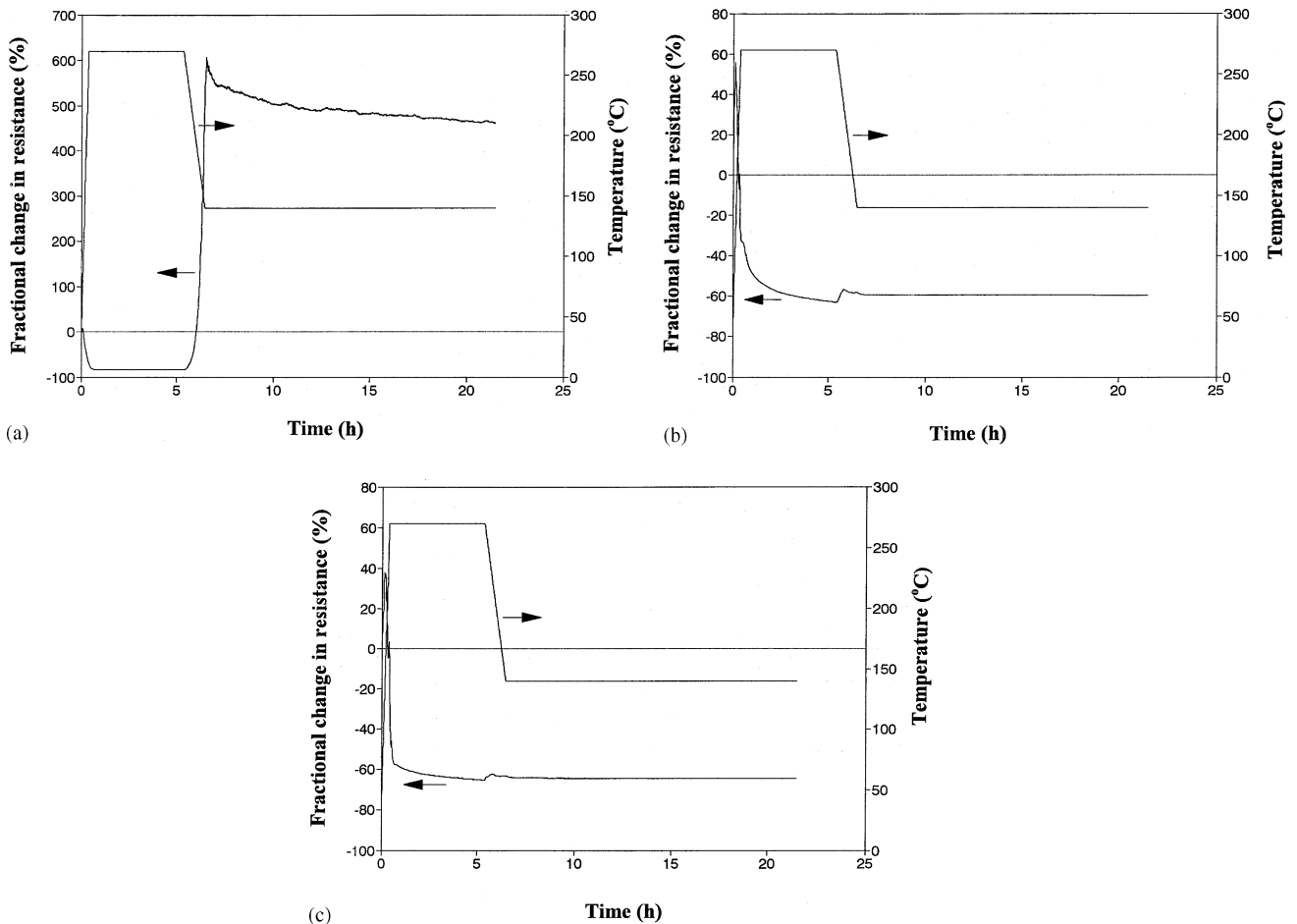

(a)

(b)

(c)

Fig. 3. The effect of bonding pressure on the contact resistance variation at a fixed bonding temperature of 270°C. (a) 4.8×10^3 Pa. (b) 3.0×10^5 Pa (c) 6.8×10^5 Pa.

This large effect allows contact electrical resistance measurement to provide nondestructive bond testing.

4. Conclusion

Thermal stress-induced debonding of PPS from PPS was studied nondestructively by measurement of the contact electrical resistance, which increases sharply upon debonding. In spite of the bond attained during bonding, debonding occurred upon subsequent cooling if either the temperature or the pressure was not sufficiently high during debonding. For PPS, the minimum bonding temperature was 290°C when the pressure was 4.8×10^3 Pa, and the minimum pressure was 3×10^5 Pa when the bonding temperature was 270°C.

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