# The Ultrasound-Based Interfacial Treatment of Aramid Fiber/Epoxy Composites

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**ABSTRACT:** The quality of interfacial adhesion of aramid/epoxy composites affects the mechanical performance of the material, and thus there is a need to improve the condition by using the ultrasound-based interfacial treatment. To do so, an ultrasonic transducer has been developed and evaluated under various operational conditions when it is installed in the winding system. It has demonstrated several key characteristics such as low power, high amplitude (more than 80 μm), and continuous working (more than 8 h) without water-cooling. Subsequently, experiments were carried out to determine the mechanical performance of the polymer material with and without ultrasound treatment, showing that the ultrasonic treatment has improved the interfacial performance up to 10%, compared with those without any ultrasound-treatment. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 113: 1816–1821, 2009

**Key words:** composites; mechanical properties; ultrasound treatment

## INTRODUCTION

Aramid/epoxy composites are widely used in military, aviation, spaceflight, industry, civil engineering structure, and so forth,[1–5] as they are immune not only to corrosion but also to electrical conduction. In addition, they demonstrate high strength, high modulus, low weight, and high fatigue strength. Some composite specimens reported by some of the authors[6] have been shown to be easily shear-destroyed, and this is usually caused by the poor wetting resistance of the material, which creates a weak interface due to short periods of wetting. There are several techniques that have been reported to improve the interfacial adhesion, and these include both the chemical and plasma treatments.[7–8] However, it is difficult to control the chemical treatment on the surface of aramid, and the process is usually time-consuming. In addition, the higher power used for the chemical treatment can result in damages in the aramid fiber. On the other hand, the improved mechanical performance of aramid after plasma treatment cannot be sustained after the treatment is removed. In light of the above, a novel ultrasonic technique is introduced to improve the interfacial adhesion of aramid/epoxy composites. The details are discussed below.

An ultrasonic transducer for the interfacial treatment was designed and constructed. It is required that the transducer be operational without water-cooling and that the resulting ultrasonic vibration system is created on the basis of a theoretical model that has characteristics such as low power consumption, high amplitude (more than 80 μm), and continuous working (more than 8 h) without water-cooling. Furthermore, the working surface of the vibration system designed should satisfy the needs of high winding speed of the fiber.

## THEORETICAL ANALYSIS OF ULTRASOUND TREATMENT ON THE INTERFACE OF FIBER AND RESIN

The mechanical strength of composites is realized through the interaction between the fiber and resin, and this can be enhanced through ultrasonic treatment. A series of tests carried out in the Mechatronics Laboratory at Harbin Institute of Technology in China showed that ultrasound can enhance the strength between the fiber and epoxy through various effects, which include cavitation, subsequent second-order effect, great mechanical effect, and temperature effect, so that at the surface of the aramid fiber, the interface of aramid/epoxy and viscosity of resin can achieve a better performance than those without. As a result of the above factors, the

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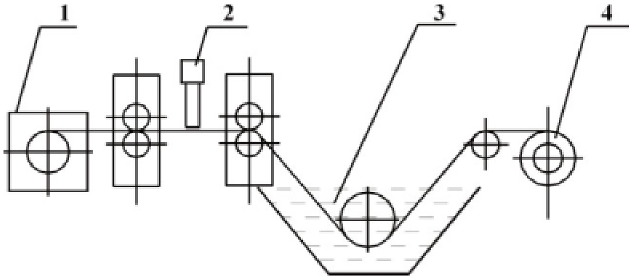

**Figure 1** Schematic of the ultrasound-based treatment system on interface of composites (1, coiler; 2, ultrasonic vibration system; 3, resin bath; 4, fiber).

wetting performance of fiber embedded in resin is improved.

### Removing gases using ultrasound

During the winding process of fiber, when the fiber embodied in resin is drawn out of the resin bath continuously, the gas impurities can easily be mixed into the resin, and the bubbles created are absorbed by the holes on the fiber surface. This effect reduces the interface strength of composites and the bubbles arisen from the impure gases form the stress points in the composites. It is important to note that the ultrasound offers an important solution to effectively remove the gases, as acoustic pressure can drive the bubbles out of the resin. In addition, the ultrasound used can decrease the viscosity of resin, which makes the bubbles mobile and thus easily removed.

### Corrosion function of ultrasound

The winding aramid fiber is exposed not only to the impurities but also to oxygen, which is easily absorbed to form a dirty surface on the fiber, causing the surface energy of fiber to be reduced dramatically compared with that of a clean fiber. This results in an incomplete contact between the aramid fiber and the resin, thus affecting the mechanical strength, especially the shear strength. When an ultrasonic treatment system is switched on, with the parameters being optimized, the acoustic cavities would oscillate, causing ultrasonic cavitation developed inside the resin. The cavities expand under the negative pressure of ultrasound and shrink under positive pressure. Under high acoustic pressure conditions, the cavities explode at an extremely high speed for an instant to create an enormous shock wave, which results in huge destruction on the dirty surface of the fiber. This characteristic is termed as the corrosion function of ultrasound, by which the wetting of fiber can be improved.

### Dispersion function of ultrasound

When ultrasound is used to treat the interface formed by the two media, fiber and resin, the acoustic steaming at a high speed arising from ultrasonic cavitation, termed the second-order effect of ultrasonic cavitation, causes the resin to disperse on the fiber surface evenly and quickly, which is called dispersion function of ultrasound. It is important to note that evenness of resin on the fiber affects the wettability performance. As a result, the fiber may accomplish good wettability performance

### Temperature effect of ultrasound

During the winding process, when the fiber impregnates into the resin, the choice of low viscosity of resin will improve wettability. When ultrasound is applied to the two media, the enormous energy of the ultrasound is focused on the interface, which is subsequently converted into heat energy. The temperature of resin rises as a result, and this is called the temperature effect of ultrasound. In summary, the temperature effect benefits the decrease of viscosity, surface tension, and surface energy, thus improving wettability.

## THE COMPONENTS OF THE ULTRASOUND SYSTEM FOR THE COMPOSITE INTERFACE TREATMENT

The winding-forming technology is used to make a specimen for tests using the NOL (spell it out) hoop. A schematic of the ultrasound-based treatment system is illustrated in Figure 1 and the experiment setup photo is shown in Figure 2, where the aramid fiber embedded in resin is drawn out of the resin bath at a certain speed, which is required to go through the two mounted squeezing rollers, and subsequently the fiber is treated by the ultrasonic transducer, excited by a generator operating at a certain frequency. It is important to point out that the frequency of the generator varies automatically with the resonant frequency of the transducer excited, termed trace-frequency. The trace-frequency is evaluated to ensure that the trace-precision is good

**Figure 2** The experiment setup photo. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

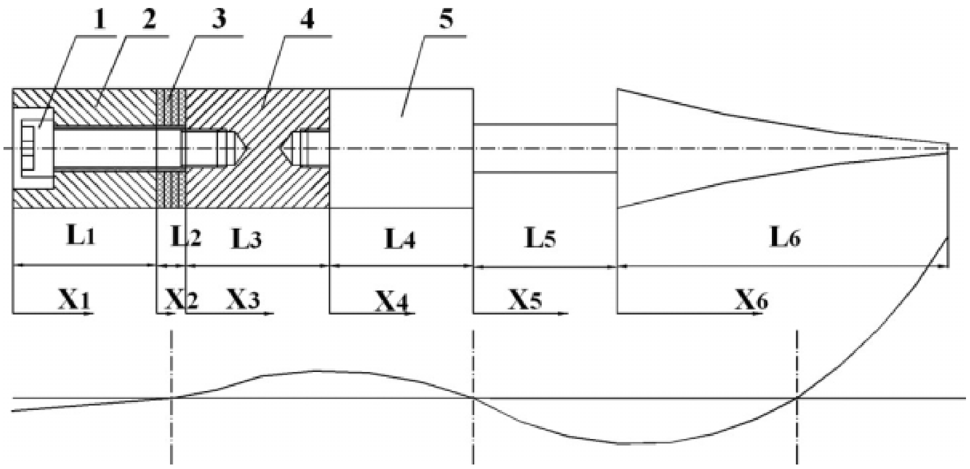

**Figure 3** Schematic diagram of the ultrasonic vibration system (1, pre-stress bolt; 2, rear plate; 3, piezo-ceramic; 4, front plate; 5, cascade horn).

enough for the application. After being heat-dried at a certain temperature for a certain period, an NOL hoop specimen for tests is created, shown in the Figure 4, and subsequently is cut into several parts for the shear-destroy tests. The main purpose of this experiment based on an NOL hoop is to evaluate the shear strength of the material after the ultrasonic treatment and to cross-compare it with those without any ultrasonic treatment.

## DESIGN OF AN ULTRASONIC VIBRATION SYSTEM FOR THE TREATMENT OF INTERFACIAL ADHESION OF COMPOSITES

The parameters of the ultrasonic vibration system are specifically chosen, based on the extensive prior research by some authors,[9,10] and it is reported that an optimum condition can be reached when a vibration system runs at a frequency of 18–22 kHz, with a signal amplitude of 30–80 μm and the treatment time of 30–40 min. However, the detailed design of the vibration system for the treatment was not discussed in the published papers; the speed of winding reported was 10 mm/min, and it was lower than the speed of 1 m/min set in this work. Based on the prior work reported, a cascaded horn is developed in this work to ensure the amplitude of 80 μm at the working surface of the horn, under continuous winding conditions without water-cooling.

In this work, the working surface of the vibration system is 4 mm × 20 mm, rectangle-shaped, which is very well suited to the high winding speed of fiber because the rectangle-shaped working surface makes the ultrasonic energy more focused on composites compared to the other shaped working surface. As a result, the vibration system functions effectively at 20 kHz and the output amplitude of the transducer operates at 80 μm.

### Design of transducer

The schematic diagram of the longitudinal piezoelectric transducer is shown in Figure 3. The dimension and material of the designed transducer are shown in Table I, where $l_1$, $l_2$, and $l_3$ satisfy the equation at a designed frequency,[11] and the amplitude magnification of the transducer is 2.6.

### Design of the cascaded horn

The cascaded horn designed in this paper is composed of the two parts, with one shape being echelon and the other exponential. The horn is made of titanium alloy because it has some advantages over

**TABLE I**
**Dimensions and Materials of the Designed Piezoelectric Transducer**

| Name | Material | Diameter (mm) | | Length (mm) |
|---|---|---|---|---|
| | | Internal diameter | External diameter | |
| Piezo-ceramic | PZT-8 | 20 | 50 | 5 |
| Front plate | Super hard aluminum | 50 | | 62 |
| Rear plate | Mild steel | 50 | | 60 |

**Figure 4** The NOL hoop specimen for tests. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

the others in terms of low acoustic impedance, high strength, and good corrosion resistance.

### Design of the echelon-shaped horn

When the amplitude node location in the echelon-shaped horn is designed at the common border between the large end and the small end of the step horn, illustrated in Figure 3, $l_4$ and $l_5$ of the step-shaped horn can be given by the propagation equation,[12] i.e., $l_4 = l_5 = 61.25$ mm, which is material dependent, as titanium alloy was chosen in this case. The boundary condition may satisfy the acoustic impedance at the amplitude node location, and thus the amplitude magnification of the step horn under the condition of non-loads can be worked out, ie,

$$MP_1 = \left|\frac{\upsilon_{e1}}{\upsilon_{f1}}\right| = 6.25$$

where $\upsilon_{f1}$ is the vibration velocity at the input end of the echelon-shaped horn, and $\upsilon_{e1}$ is the vibration velocity at the output end of the step-shaped horn.

### Design of the exponential-shaped horn

The input end of the exponential-shaped horn is 45.96-mm long and 45.96-mm wide, i.e., $a_1 = 45.96$ mm, $b_1 = 45.96$ mm. The area value of the input end is $s_1 = a_1 \times b_1$. The output end of the exponential-shaped horn is 20-mm long and 4-mm wide, i.e., $a_2 = 20$ mm, $b_2 = 4$ mm. The area value of the output end is $s_2 = a_2 \times b_2$, which satisfies the equations below:

$$a_2 = a_1 \cdot e^{-2\beta_1 l_6}$$

$$b_2 = b_1 \cdot e^{-2\beta_2 l_6}$$

$$s_2 = a_2 \cdot b_2 = s_1 e^{-2(\beta_1+\beta_2)l_6} = s_1 e^{-2\beta l_6}$$

where $\beta = \beta_1 + \beta_2$, $\beta_1$ and $\beta_2$ are the decay constants of the length and of the width, respectively, and $\beta$ is the decay constant of area. The resonance length of the exponential-shaped horn is $l_6 = 138$ mm. The coordinate of the amplitude node in the exponential-shaped horn is $x_6 = 47.95$ mm. The amplitude magnification of the exponential-shaped horn is $MP_2 = 5.138$. So the amplitude magnification of the cascade-shaped horn is:

$$MP = MP_1 \times MP_2 = 32$$

which is well accorded with demands.

## EXPERIMENTS

To study the effects of the ultrasonic treatment on interfacial property between aramid and resin, a series of tests on the composites was carried out, these being achieved under the conditions in which different ultrasonic amplitudes were generated and the results obtained are cross-compared with those from the non-ultrasound treatment. The materials are made of epoxy, and acid anhydride is used as curing agent to wind the NOL hoop specimen at a speed of 1 m/min, shown in Figure 4. All the reported and experimentally obtained values of the interfacial shear strength are analyzed and evaluated in relation to the GB1416-88 standard. In this work, five NOL hoop specimens were wound under the same ultrasonic conditions, with each NOL hoop specimen being cut to eight parts for shear strength tests. During the process of winding, human error made the results disperse to some extent. Therefore, a statistical method was introduced to evaluate the data obtained. The dispersion coefficient $V_p$ (%) is given below

**TABLE II**
**Shear Performance of Composites Treated at Different Ultrasonic Amplitudes**

| | Untreated by ultrasound | Ultrasonic amplitudes (μm) | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 29 | 34 | 40 | 48 | 56 | 68 | 78 |
| Average of shear strength (MPa) | 48.1 | 48.86 | 49.6 | 52.1 | 52.9 | 50.95 | 49.1 | 37.5 |
| Range of shear strength (%) | – | 1.58 | 3.11 | 8.31 | 9.97 | 2.850 | 2.07 | −22.03 |
| Disperse coefficient (%) | 3.54 | 2.21 | 2.51 | 2.07 | 1.57 | 4.08 | 5.66 | 6.2 |

$$V_p = \frac{x_{i\max} - x_{i\min}}{x_i}$$

where $x_{i\max}$ is the maximum value, $x_{i\min}$ is the minimum value, and $x_i$ is the average value of shear strength at certain ultrasonic amplitudes respectively.

To show the improvement of shear strength in percentage compared with the value of shear strength without the ultrasonic treatment, the data obtained from the ultrasound treatment have been evaluated by the relative of shear strength, $\Delta T/T$, which is given by the equation below.

$$\frac{\Delta T}{T} = \frac{x_i - x}{x}$$

where $x$ is the value of shear strength without the ultrasonic treatment, and $\Delta T/T$ is range of shear strength (%).

The reported values shown in Table II are the statistical values of shear strength, range of shear strength, and of the corresponding dispersion coefficient when the winding speed is 1 m/min.

Table II shows that the shear performance was significantly improved by 10% when the dispersion coefficient was 1.57%. These values were obtained at the ultrasonic amplitude of 48 μm.

Scanning electron microscope (SEM) photos of the destructive NOL hoop specimen are shown in Figure 5, obtained under conditions when the ultrasonic treatment amplitude varies. Images in Figure 5(b,c) were obtained when the ultrasonic amplitude was 48 μm and of 78 μm, respectively; in Figure 5(a), there was no treatment. As shown in Figure 5, the surface of the fiber was covered by little resin, and thus the shear performance was decreased due to the wettability performance degradation. The result shown in Figure 5(b), obtained at the ultrasonic amplitude of 48 μm, achieved the best performance over the range from 29 to 78 μm both in terms of the shear performance and of the wettability, with which the stability of composites tends to increase. The fiber shown in Figure 5(c) is subjected to the ultrasonic treatment at amplitude of 78 μm, and it shows that the interface adhesion has been destroyed totally by the cavitation arisen from ultrasound, which causes the resin being difficult to adhere to, this being nearly the same as that from Figure 5(a).

The experimental results obtained are in good agreement with those obtained from the theoretical discussions. Experiments carried out demonstrated that the wetting performance of fiber embedded in resin was improved, with which both the interfacial adhesion strength between the fiber and resin and the mechanical performance of composites improved accordingly.

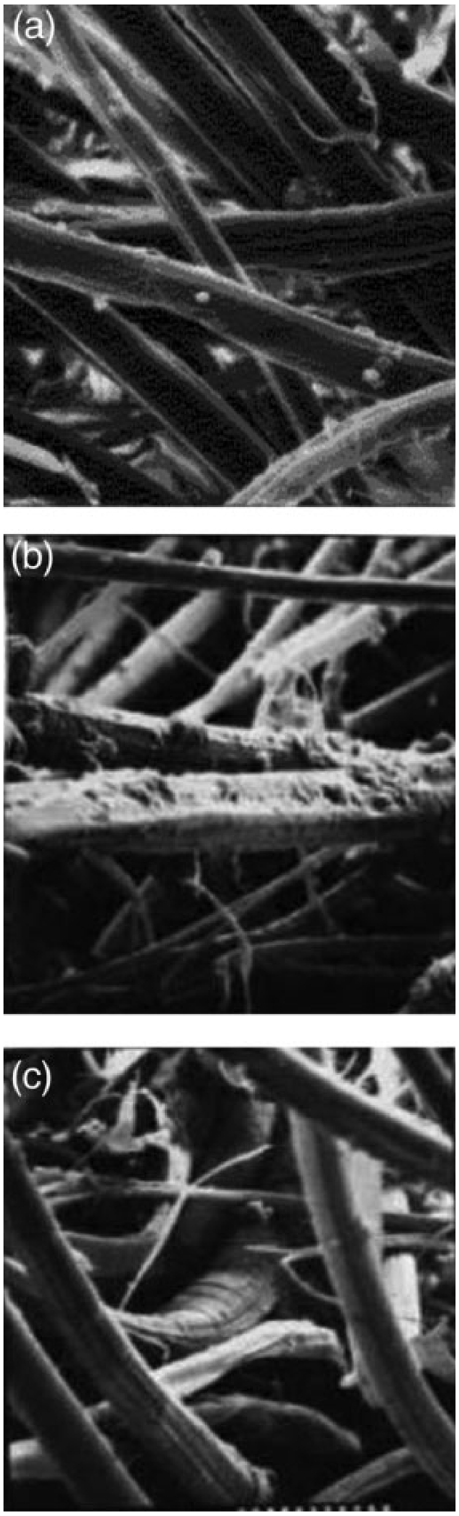

**Figure 5** Scanning electron microscope photos of destructive NOL hoop specimen (a) under the condition of the nonultrasound treatment; (b) at ultrasonic amplitude of 48 μm; and (c) at ultrasonic amplitude of 78 μm.

## CONCLUSION

A novel ultrasound-based polymer winding equipment has been specifically set up and evaluated in this work to evaluate and determine the interfacial characteristics between aramid and resin. The experimental results obtained have confirmed that the shear strength of polymer was improved by 10% when treated by ultrasound with an optimized setting, compared with those without the ultrasound treatment.

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