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Flame and silane treatments for improving the adhesive bonding characteristics of aramid/epoxy composites

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ABSTRACT

A fast and cost-effective surface treatment with flame and silane coupling agent treatments has been developed for the surface treatment of aramid/epoxy composite faces to improve the adhesive bonding characteristics of lightweight sandwich stealth radome structures. The flame treatment was performed with propane gas, and the silane treatment was performed with γ-methacryloxypropyltrimethoxy silane (γ-MPS) and γ-aminopropyltriethoxy silane (γ-APS) under different treatment conditions. The contact angles of the flame-treated aramid/epoxy composites and the single-lap shear strengths of the adhesive joint composed of the aramid/epoxy composite adherend and epoxy adhesive were measured and compared with those treated with argon plasma and mechanical abrasion. In addition, the surfaces were analyzed with atomic force microscopy (AFM), scanning electron microscopy (SEM) and X-ray photoelectron spectroscopy (XPS) to characterize the mechanical and chemical interactions of the aramid/epoxy composite with the epoxy adhesive.

1. Introduction

Structures made of aramid-fiber-reinforced composites have been widely used in aerospace structures and ship constructions due to their high specific strengths [1]. The main function of sandwich radome structures in marine applications is to protect indoor radars from external environments such as heat, moisture and dynamic impacts. The sandwich stealth radome structure, composed of composite faces, frequency selective surfaces (FSS) and foam, has suitable mechanical and dielectric properties [2]. In order to fabricate stealth radome structures, a reliable set of adhesive bonding characteristics between the radome components without any defects will increase the radar cross-section and is indispensable. In this work, the aramid/epoxy composite was selected as the load-bearing face structure of the sandwich radome because of its high specific mechanical properties and low level of transmission loss for EM (electromagnetic) waves.

Many studies have investigated improving the adhesive bonding characteristics of aramid composites with treatments such as silane treatments, plasma surface treatments and mechanical abrasion. For the silane treatments, γ-methacryloxypropyltrimethoxy silane (γ-MPS) is generally used to optimize and promote the adhesion through chemical and physical coupling between composite–composite, metal–composite and ceramic–composite materials. Any composite material that contains methacrylate groups in the molecules can be treated with MPS because the methacrylate end group copolymerizes better to the composite than acrylate [3]. The compound γ-aminopropyltriethoxy silane (γ-APS) is used to treat aramid fibers or composites for strong interfacial interaction between the matrix and fibrous reinforcement [4]. Boerio et al. have found that amino silane formed hydrolyzed polysiloxanes that are weakly bound and easily desorbed by water. It was suggested that the amino silane would be adsorbed with the electron pair on the fiber surface of organic materials that had a non-polar chemical structure [5]. Yue and Quek reported that the silane coupling agent containing an amino silane coupling agent improved the mechanical interfacial properties of composites, as compared to a single coupling agent [6]. It is generally well-known that organic functional silanes are the most widely used coupling agents in glass-fiber-reinforced epoxy resin composites because the silane coupling agent can easily react between inorganic and organic materials simultaneously, and the adhesion performance of the glass/epoxy composites can be effectively improved [7,8].

However, there are few studies on silane coupling agents that analyzed aramid/epoxy composite surfaces for improving the chemical interaction and the bonding characteristics of the composite for increasing the adhesion strength of the adhesive joints.

In this study, the aramid/epoxy composite faces were flame-treated with propane gas; silane treatments were performed on the flame-treated surfaces to further improve the bonding

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performance. The silane coating treatments were performed on the surface of flame-treated aramid/epoxy composites with γ-MPS and γ-APS. The surface free energy was calculated by measuring the contact angles on the aramid/epoxy composite adherend with respect to the weight concentration of the silane solution, which was used to investigate the relationship between the surface free energy and the single-lap shear strength of the aramid/epoxy composite joint. A quantitative chemical bonding analysis with X-ray photoelectron spectroscopy (XPS) was also conducted to observe the chemical binding states of the surfaces of the aramid/epoxy composite with respect to the surface treatment condition. A suitable flame treatment condition for the aramid/epoxy composite surfaces was suggested to maximize coupling effects between the aramid/epoxy composites and the epoxy adhesive without surface degradation.

From the experimental results, the optimum flame and silane treatment conditions for the enhancement of the chemical interaction of silane coupling agents were obtained. It was found that the flame- and silane-treated composite surface greatly improved the adhesion strength of aramid/epoxy composite joints under the optimum treatment condition due to the increased number of oxidized radicals on the composite surfaces.

2. Experimental

2.1. Specimen preparation and experimental set up

Adhesively bonded single-lap joints composed of the aramid/epoxy composite adherends and epoxy adhesive were fabricated as shown in Fig. 1. Table 1 shows the mechanical properties of the aramid/epoxy composite (Heracron 1000d, Kolon, Korea). The adhesively bonded joint bonded with an epoxy adhesive (Araldite, Hutsman, England) whose material properties were shown in Table 2, was cured at 60 °C for four hours. The thickness of the adhesive layer was fixed at 0.1 mm by using a spacer mold for the maximum joint strength [9]. After curing the adhesively bonded joint, the adhesive fillets of the joint were removed with a razor blade to reduce the variation in lap shear strength. Then, the lap shear strengths of the aramid/epoxy composite joints were measured on a computer-controlled INSTRON static 4206 material testing system (INSTRON, USA). The lap shear tests were performed with a loading speed of 1.0 mm/min at the test temperature of 25 °C. From the measured static tensile load capacity F, the arithmetic lap shear strengths P were calculated as follows:

$$P = \frac{F}{wl} \tag{1}$$

where w and l are the bond width (20 mm) and length (20 mm) of the lap shear joint, respectively. At least five specimens were tested for each type of the aramid/epoxy composite to check for repeatability.

2.2. Surface treatments of the aramid/epoxy composite

2.2.1. Flame treatment

Flame treatment using propane (C_3H_8) gas was used to modify the surfaces of the aramid/epoxy composite and to eliminate surface contamination. Fig. 2 shows a flame burner for liquefied petroleum gas (LPG) for flame treatment of the aramid/epoxy composite. The nozzle diameter and length of the flame burner was 1.0 mm and 600 mm, respectively. The flame treatment was performed under a pressure of 25 kPa with a treatment distance of 20 mm. The surface temperature of the flame was about 900 °C. Perfect combustion could be achieved using a gas regulator for the flame burner during the flame treatment. The surface treatment was performed on the aramid/epoxy composite surfaces for different treatment times to investigate the effects of flame treatment on aramid/epoxy composite surfaces.

2.2.2. Silane treatment

The chemical structures of the silane coupling agents, γ-MPS and γ-APS (Shinetsu, Korea), are shown in Fig. 3. To treat the aramid/epoxy composite surface under different weight concentrations of silane solution, the γ-MPS and γ-APS concentrations were varied from 0.1 to 5 wt.% in distilled water, which are the treatment ranges recommended by the manufacturer of the silane solutions. The silane treatment process for the single-lap shear test is shown in Fig. 4. For the silane treatment on aramid/epoxy composites, an acidic solution was prepared by mixing distilled water and acetic acid (CH_3COOH) to a pH of 4.5. Then, the silane coupling agents were mixed with the acidic solution with a magnetic stirrer at a rotating speed of 500 rpm for 60 min until the silane solution became transparent, which indicates the completion of hydrolysis of the silane solution. The aramid/epoxy composites were immersed into the hydrolyzed silane solution for 3 min at 25 °C, which saturated the adsorption of the silane solution on the aramid/epoxy composite surfaces. Finally, the silane-treated aramid/epoxy composites were dried at 60 °C for 2 h in an oven to promote the condensation of silanols to siloxanes.

2.3. Analysis of treated surfaces

2.3.1. Atomic force microscopy (AFM) and scanning electron microscopy (SEM) analysis

The surface characteristics of the aramid/epoxy composite were examined with an atomic force microscope (AFM) (XE-100, Park Systems, Korea) at ambient conditions. The scan size was 1 μm × 1 μm. All the images had 256 data points, and a scan rate of 1.0 Hz was used. The arithmetic mean surface roughness was measured for different treatment times. After the treated surfaces of the aramid/epoxy composite sample were vacuum-coated by evaporation with gold before examination, they were analyzed with a scanning electron microscope (SEM) (XL30SFEG, Philips,

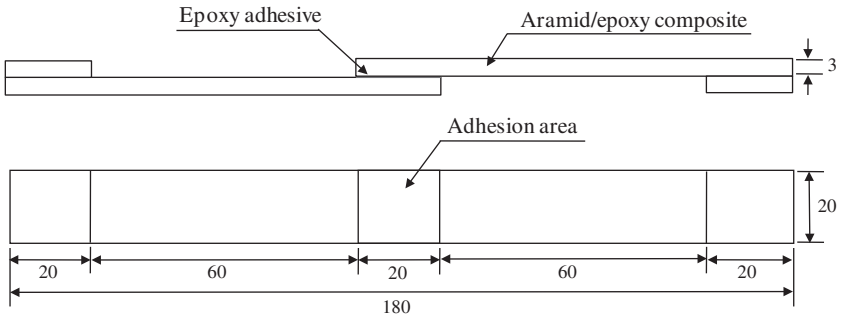

Fig. 1. Schematic drawing of the single-lap joint specimen (dimensions in mm).