



Effect of surface treatment on the interfacial adhesion performance of aluminum foil/CFRP laminates for cryogenic propellant tanks

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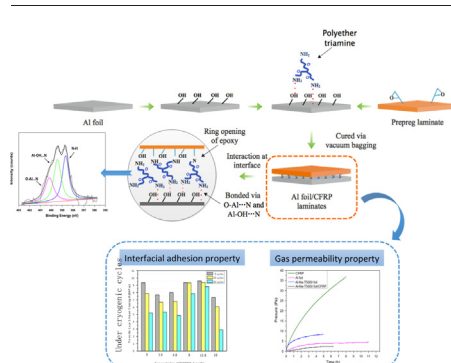
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HIGHLIGHTS

- Al foil was inserted between plies of carbon fiber reinforced polymer to manufacture laminates.
- Cryogenic cycling and tensile lap-shear test were combined to measure the interfacial performance.
- Corrosion methods could improve the tensile lap-shear strength by 96%.
- Flexible layer could enhance the interfacial adhesion performance after cryogenic cycles significantly.
- The laminates exhibit superior gas barrier property to that without Al foil.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, an aluminum foil with a thickness of 0.08 mm was inserted between plies of carbon fiber reinforced polymer (CFRP) composites to manufacture Al foil/CFRP laminates. Two kinds of surface treatments, including corrosion method with NaOH or H₂SO₄/CrO₃ solution and addition of flexible layer composed of polyether triamine, were carried out successively on the surface of Al foil to improve the interfacial adhesion strength between Al foil and CFRP under cryogenic cycles. The results showed that, both corrosion methods could improve the interfacial adhesion strength between Al foil and CFRP obviously. The flexible layer applied was verified to be bonded to the surface of Al foil via O—Al...N and Al—OH...N. The interfacial adhesion performance of Al foil/CFRP laminates after cryogenic cycles improved significantly attributable to the surface treatments applied, which indicated that the flexible layer could adjust the thermal expansion coefficient mismatch between Al foil and CFRP as expected. Furthermore, the Al foil/CFRP laminates exhibit excellent gas barrier property, which is about 15 times higher than that of CFRP laminates. Consequently, the Al foil/CFRP laminates have great potential as cryogenic propellant tank material for aerospace craft.

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1. Introduction

Drastic weight reduction and recyclability of propellant tanks are needed for the realization of aerospace craft with increasing carrier capability as well as decreasing cost, such as reusable launch vehicles (RLVs) [1–4]. As propellants are usually cryogenic media, traditional

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propellant tanks are metal tanks, which are able to meet the requirements of mechanical and barrier properties under cryogenic temperature and have a mature manufacture technology. Nevertheless, isotropic metal material could not meet the demand for weight reduction of future aerospace craft [1,5].

Thus, it is imperative to develop an alternative material for cryogenic tanks. In 1987, the first attempt of manufacturing liquid hydrogen (LH₂) tank with carbon fiber reinforced polymer (CFRP) composites was completed by McDonnell Douglas Aerospace (MDA) in USA [6]. Thereafter, enormous interests have been attracted in designing and manufacturing CFRP cryogenic propellant tanks. Attribute to the drastic lightweight, high specific mechanical performance and structure/function designability, CFRP has become the most promising material for aerospace propellant tanks [7–9]. However, cryogenic composite tanks still have not been maturely applied up to now. One of the major challenges is the propellant (gas and liquid) leakage, which occurs through microscopic damage, such as cracks in matrix, fiber/matrix debonding, and delamination [10–13].

In order to prevent the propellant leakage through the wall of CFRP, metal material, such as aluminum alloy, titanium alloy or stainless steel, was usually used as a structural or non-structural gas-tight shell inside the CFRP body, which is called metal-lined CFRP cryogenic tank [14–15]. A lot of corresponding researches have been carried out, including flight test [15], damage analysis [16], non-destructive inspection [17], etc. The results showed that, although the addition of metal liner could prevent the gas diffusion and propellant leakage under cryogenic conditions effectively, debonding on the interface of metal and CFRP occurred often resulting in shorten life and insecurity of the tanks and aerospace craft. Furthermore, as being the core mold in fiber winding molding, metal liners used in preceding works had a certain thickness of about 2–3 mm [15,17,19], which limited the weight reduction of the cryogenic propellant tanks.

In recent years, metal foil inserted CFRP laminates have been developed, which could retain the excellent barrier property of metal-lined CFRP tanks while achieve the maximum weight reduction [18]. However, debonding on the interface between metal and CFRP is still inevitable. In addition to the plastic deformation due to different stiffness between metal and composites [19–20], the severe thermal stress attributable to the coefficient of thermal expansion (CTE) mismatch between metal and composites is a non-negligible reason, which becomes much greater at cryogenic temperature than at room temperature (RT).

Concerning the improvement of bonding strength between metal and CFRP, several methods have been proposed. Sun et al. [21] demonstrated that randomly-distributed short fibers could toughen the interface of CFRP and aluminum substrate by increasing the energy release rate (G_c) from 10 J/m² to 335 J/m². Takeuchi et al. [22] introduced a slit in the mouthpiece near the bonding layer between composite and metal, which strengthened the bonding structure by reducing the energy-release rate to less than one-tenth of its original value. Likewise, Mei et al. [23] modified the surface of aluminum with one dendrimer, aiming to enhance the interfacial adhesion strength between metal and CFRP. However, the interfacial adhesion performance of metal/CFRP laminates under cryogenic cycles has not been investigated. The cryogenic cycling is a simulation of the actual service condition of reusable cryogenic tanks.

In the current work, considering barrier property and further weight reduction, aluminum foil with a thickness of 0.08 mm was utilized for manufacturing Al foil/CFRP laminates. Instead of being a liner, the Al

foil was inserted in the composite material. In order to improve the interfacial adhesion between Al foil and CFRP, Al foil was corroded firstly. Thereafter, a flexible layer of polyether triamine, which has been approved to have excellent cryogenic impact toughness [24], was fabricated on the interface to adjust the CTE mismatch between two substrates. The effects of surface treatments on surface functional groups, morphology and wettability were evaluated in detail. The adhesion properties, with and without surface treatments, were compared based on tensile lap-shear strength after several cryogenic cycles. Furthermore, the gas barrier properties of Al foil/CFRP laminates were investigated. This design of Al foil/CFRP laminates aimed to fabricate a material with not only improved interfacial adhesion performance under cryogenic cycles but also excellent gas barrier property, so as to provide technical support for the application of lightweight, high-barrier and recyclable cryogenic propellant tanks.

2. Experimental

2.1. Materials

The carbon fiber reinforced epoxy resin prepreg employed in this research was supplied by CASIC (Beijing, China), which is comprised of epoxy resin matrix and unidirectional T700 carbon fiber reinforcement. The aluminum foil, having a thickness of 0.08 mm, was provided by Tong Fatai Trade Co., Ltd. (Tianjin, China). Polyether triamine (Jeffamine® T5000), with weight-average molecular weight of 5000, was obtained from Huntsman Corporation (Texas, USA). NaOH, K₂Cr₂O₇, H₂SO₄ and acetone were all purchased from Beijing Chemical Works (Beijing, China).

2.2. Sample preparation

2.2.1. Surface corrosion of aluminum foil

Aluminum foils were first cleaned ultrasonically in acetone for 5 min and dried at 50 °C for 1 h. The so-cleaned aluminum foil is referred to as the pristine Al foil (shorthand for Al foil). As proper surface corrosion is essential for successful bonding, Al foils were corroded with sodium hydroxide (NaOH) solution and sulfuric-chromic acid (H₂SO₄/CrO₃) solution respectively. The specific processes are presented in Table 1.

2.2.2. Preparation of flexible layer

Polyether triamine T5000 was first dissolved in acetone and well stirred, then the Na⁺-Al foil (Al foil pretreated with NaOH solution) was dipped in the T5000 solution at 25 °C for 1 h; at last, the treated Al foil (T5000-Na⁺-Al foil) was dried at 50 °C for 1 h.

2.2.3. Manufacture of Al foil/CFRP laminates

The CFRP laminate for tensile lap-shear test was comprised of 3 or 4 plies of prepreg measuring 180 mm × 100 mm with the fibers in the 0° direction paralleling to the 100 mm side. For each specimen, two pieces of the above laminates were required. The Al foil was measured 180 mm × 60 mm and placed on the surface of one CFRP laminate, followed by lapping with the other piece of CFRP laminate. The lap form, dimensions and alignment of the specimens were shown in Fig. 1.

For tensile and flexural tests, 5 and 15 plies of prepreg measuring 250 mm × 60 mm and 120 mm × 13 mm were laminated respectively to produce CFRP specimens. As for the Al foil/CFRP laminates, one piece of Al foil was inserted into the middle of the CFRP laminates prepared above. The fibers in the 0° direction were parallel to the long side.

Table 1
Methods for surface corrosion of Al foil.

Sample	Agent	Concentration	Method
Na ⁺ -Al foil	NaOH	50 g/L	Dipped in NaOH solution at 65 °C for 0.5–2.0 min, rinsed with deionized water, dried at 50 °C for 1 h.
Cr ³⁺ -Al foil	H ₂ SO ₄ /CrO ₃	m(H ₂ SO ₄):m(K ₂ Cr ₂ O ₇) = 10:1	Dipped in H ₂ SO ₄ /K ₂ Cr ₂ O ₇ solution at 65 °C for 10.0 min, rinsed with deionized water, dried at 50 °C for 1 h.

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