Enhancing interfacial bonding strength in fiber metal laminates through metal surface treatments

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Abstract In recent decades, the aircraft industry has experienced a growing need for high-performance, lightweight structures, leading to a significant focus on developing fiber-metal laminates (FMLs). These composites possess various benefits in terms of stiffness, yield stress, fatigue, and high-velocity impact properties due to their hybrid composition. However, certain challenges related to the fabrication, surface treatment, and mechanical properties of these structures require further attention. This paper reviews FML manufacturing with a focus on enhancing the interfacial bonding strength between metal and fiber-reinforced polymer (FRP) sheets by conducting different kinds of surface treatment on metal layers to further enhance the FML mechanical properties.

Introduction
The growing demand in the transportation industry for reduced fuel usage and subsequent decrease in CO₂ emissions is driving the development of structural materials having a lightweight still with enough strength. FMLs are composite materials combining metal and FRP sheets, as depicted in Fig.1. Originating from the aviation sector with the primary aim of enhancing fatigue resistance, FMLs have emerged as a highly desirable structural option due to their exceptional strength-to-weight ratio [1].

![Fig. 1 Configuration of different FMLs: 2/1, 3/2, and 4/3 lay-ups [2].](image)

Aluminum alloy-based FMLs are extensively used in various industrial applications. The most often used types are aramid fiber-reinforced aluminum laminates (ARALL), carbon fiber-referenced aluminum laminates (CARALL), and glass fiber-referenced aluminum laminates (GLARE) [3]. The growing need for industrial application of lightweight and improved mechanical and thermal properties has led to the development of FMLs based on magnesium and titanium alloys to fulfill these requirements. Thermoset polymers have been extensively utilized...
as the matrix for CFRPs in the composite core [4-5]. However, the advantages offered by thermoplastic polymers, including decreased cycle time, recyclability, and improved compatibility with existing stamping processes [6], are positioning them as viable alternatives to thermoset ones. As a result, they have gained significant attention from both academic and industrial researchers.

Nevertheless, the utilization of these materials is constrained by several drawbacks. An important issue in the production of FMLs is the long-term strength of the connection between the FRP and metal sheets. Delamination, a primary defect affecting the service lifespan of FML components, significantly impacts their mechanical properties [7]. The phenomenon arises when the bonding strength between the metal and FRP layers is insufficient under various loading situations, such as impact, penetration, blast loading, and drilling processes [8,9]. Consequently, before the fabrication of FMLs, various pretreatments are typically undertaken to enhance the interfacial bonding strength to avoid delamination defects [10] and further enhance the impact [11] and fatigue [12] resistance of the formed components. This paper aims to offer a review of the surface treatment technologies carried out on metal sheets for improving the interfacial bonding strength of FMLs.

**Metal surface treatment methods**

Several investigations on the delamination behavior of FMLs focus on various techniques to enhance the interfacial bonding strength of FMLs, primarily through pre-treatments applied to metal sheets. Three main categories of surface treatments are commonly utilized for metal sheets: (i) mechanical treatments such as abrasion [13], sandblasting [14,15] and cutting [16,17], (ii) chemical treatments including anodizing [18,19], etching [20,21], and micro-arc oxidation (MAO) [22,23] (iii) addition of interfacial layer: plasma-spray [24,25] and sol/gel methods [26,27]. The subsequent sections provide comprehensive explanations of these methods.

Mechanical treatment. Mechanical abrasion refers to the process of removing material from a surface through physical friction. This method typically involves the use of abrasive materials such as sandpaper, grinding wheels, or wire brushes, which are applied to the surface with varying levels of pressure to achieve the desired result. It can effectively remove the nonuniform oxide layer on the metal surfaces and change their surface texture. The impact of the surface morphology of the 2024-T3 aluminum alloy-based GLARE was experimentally investigated by employing sandpapers of varying grit grades (#2000, #220, and #100) to achieve different levels of roughness [13]. It was discovered that as the surface roughness increases, so does the surface energy and bonding strength, resulting in significant improvement.

Sandblasting on metal sheets is a common and effective technique for various applications, primarily in the field of surface preparation for large-scale components. When applied in the pretreatment on the metal surface of FMLs sandblasting creates a roughened and clean surface on the metal layers and enhanced surface texture promotes better adhesion between the metal and the adjacent FRP layers. Depending on the specific requirements of the FML application, the degree of roughness can be adjusted by varying the abrasive material and the blasting parameters. The effect of the change in metal surface roughness through sandblasting at different pressures was examined in [14]. Before the fabrication of FMLs via hot pressing, the surfaces of the magnesium alloy sheets underwent sandblasting at various pressures: 1, 3, and 5 bar with Fig.2 illustrating the resulting surface roughness. Metal surface roughness rises as the increase in sandblasting pressure. Interfacial bonding strength along the normal and shear directions was experimentally tested by conducting normal separation tests and single lap shear tests. The study revealed that the bonding strength exhibited an upward trend with the increasing roughness of the metal sheets.
Fig. 2 Surface roughness of the AZ31B sheets at different pretreatments [14]

While these processes effectively enhance metal surface roughness, it's common to apply additional chemical or electrochemical treatments following grit or sandblasting to achieve a stronger bond. They also investigated how the annealing process conducted on the sandblasted AZ31B sheets affects the interfacial bonding strength as Fig. 3 shows the surface morphology after sandblasting and annealing. The study observed the formation of an oxide layer, primarily composed of MgO/Mg(OH)$_2$, on the surface of AZ31B, which enhanced the metal surface roughness and facilitated chemical bonding with the polymeric molecules in the matrix. Consequently, the annealing process after sandblasting was found to further enhance the bonding strength.

Fig. 3 SEM images of the sandblasted and annealed AZ31B sheet surfaces [14]

The effective bonding area and mechanical interlocking between the metal and FRP layers also can be improved on a macro scale by producing scratches or grooves on metal surfaces. Several patterns of grooves with 0.1mm depth were cut on the surface of AA6082 T6 aluminum substrates in [16] as shown in Fig. 4, the results illustrate that 45°, 45°-crossed, 90° and 90°-crossed patterns can enhance the shear joint strength by 33% 45% 27% 42% respectively compared to the non-treated substrates.
Fig. 4 Different patterns with a depth of 0.1 mm. (a) 0° pattern; (b) 45° pattern; (c) 45° crossed pattern; (d) 90° pattern and (e) 90° crossed pattern. [16]

Similarly, grooves are wire cut on dual-phase steel DP980 surfaces as shown in Fig. 5 to strengthen the shear joint strength between steel sheets and CFRP of the FMLs fabricated through thermoforming process. The results show that the strength of the grooved specimens is more sensitive to the initial pressure compared with the grit-blasted specimens [17].

Fig. 5 Design of the grooves on DP980 surface [17].

Chemical treatment. Anodization, a method employed for treating metal sheets, alters the surface microstructure to facilitate the formation of an oxide layer, which enhances wettability, consequently improving interfacial bonding strength [28]. Various chemical combinations are usually utilized in the anodization process, including chromic acid anodization and hydroxide anodization [29]. The influence of anodizing temperature on the microstructure, composition, and surface profile of Ti6Al4V was experimentally studied in [19], with Fig. 6 depicting the SEM images of Ti6Al4V surfaces after anodization for 15 min at different temperatures. Observations revealed that at 40°C, a uniform honeycomb-like oxide layer with pore diameters ranging from 100 to 200 nm developed on the surface of Ti6Al4V. Comparatively, the apparent shear strengths of specimens anodized at 0°C, 25°C, and 40°C exhibited enhancements of 217.7%, 225.0%, and 317.2% respectively, compared to those without anodization.
Another approach to enhance surface roughness involves etching metal sheets with diverse acid mixtures. The degree of surface roughness is mainly dictated by the concentration levels of sodium hydroxide and hydrogen peroxide. Muti-step surface treatment namely sandblasting, anodizing, etching, and annealing was conducted on Ti6Al4V titanium alloy in [20]. The NaOH etching of the anodized sample significantly altered the surface morphology of the titanium alloy by producing nano-structured bumps that can effectively improve the bonding strength and wettability.

Micro arc oxidation (MAO) is a chemical treatment technique that can uniformly create a ceramic-like coating on the surface of metal sheets by generating sparks at high voltages. The metal surface treated with MAO forms an oxide film consisting of two layers: an exterior layer that is loose and porous, and an interior layer that is thick and dense. The outer porous layer facilitates interlocking with the resin, thereby enhancing bonding strength [22]. SEM surface morphology images of the titanium alloy after MAO surface treatment at various anodic voltages are depicted in Fig. 7. It is evident that an increase in anodic voltage leads to the formation of a thicker outer porous layer on the surface. With the anodic voltage elevated from 400 V to 600 V, the pore size escalates from 2 μm to 10 μm, enhancing surface wettability and resulting in stronger mechanical interlocking.

**Fig. 6** SEM images of Ti6Al4V surfaces after anodization for 15 min at different temperatures: (a and b) 0 ℃, (c and d) 25 ℃, (e and f) 40 ℃ [19]

**Fig. 7** SEM images of Ti-MAO-400 (a), Ti-MAO-500 (b), Ti-MAO-600 (c), Ti-MAO-Step at 400, 500, and 600 V [22]
Addition of interfacial layer. Plasma spraying surface treatment is a metal treatment procedure where high-pressure molten chemical powder is blasted onto the metal surface. The experimental investigation on the effect of plasma surface modification on aluminum alloy AA6061-T6 using different working gases, namely oxygen and nitrogen plasma treatment was researched in [24]. When the aluminum alloy sheet was treated with nitrogen plasma for the same duration as the oxygen plasma treatment, it resulted in a 13.7% increase in average peeling strength, a 9.4% increase in interlaminar shear strength, an 8.5% increase in tensile strength, and a 2.5% increase in flexural strength of the laminates. Another approach involves the fabrication of functional silane coatings on metal surfaces using the sol-gel technique. The sol-gel coatings consist of alkoxy groups capable of bonding with inorganic materials and functional groups that can interact with polymers. These groups can enhance adhesion on the metal-polymer interface of FMLs through the formation of covalent bonds [27].

Conclusions
The mechanical properties of FMLs are significantly affected by interfacial bonding strength. Therefore, pretreatments should be applied on metal surfaces before their fabrication to achieve a durable bond between FRP and metal alloys. Furthermore, a suitable combination of different kinds working as a multistep method can further improve metal/FRP interfacial bonding strength.

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