Chapter 9 An Overview of the Natural/Synthetic Fibre-Reinforced Metal-Composite Sandwich Structures for Potential Applications in Aerospace Sectors

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1 Introduction

A signifcant milestone has been achieved in the feld of composite materials over the past few decades. The technological evolution has ignited the development of advanced composite sandwich materials, namely, fbre-metal laminates (FMLs), in the aerospace sectors. The initial intention of developing FMLs was to tackle the poor fatigue crack resistance of metal alloys in the aerospace sectors. Later, it was found that the coalescence of metal and composite layers results in excellent impact properties with a signifcant weight reduction. The reduction in the fatigue crack growth rate and the improvement in the impact properties of FMLs have made these materials the successor over their individual constituents. In fact, the search for high damage tolerance and toughness materials for aircraft structures is motivated by the new design philosophy as introduced by the Federal Aviation Administration (FAA) (Asundi & Choi, [1997\)](#page-14-0). Therefore, the development of FMLs aims to meet the design philosophy as introduced by the FAA.

FMLs are in the category of composite sandwich laminates consisting of alternating layers of metal alloys and composites. The typical metal layers employed in FMLs are aluminium, titanium and magnesium. In addition to the excellent fatigue and impact properties of FMLs, the introduction of composite materials in the FMLs has led to weight saving. The weight reduction is particularly attractive to the aerospace sector as it could improve energy effciency. The sandwich concept was

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[©] The Author(s), under exclusive license to Springer Nature Switzerland AG 2022 177 N. Mazlan et al. (eds.), *Advanced Composites in Aerospace Engineering Applications*, [https://doi.org/10.1007/978-3-030-88192-4_9](https://doi.org/10.1007/978-3-030-88192-4_9#DOI)

initialised at the Delft University of Technology in 1978 (Mohammed et al., [2018\)](#page-16-0). The frst successfully developed FMLs were aramid fbre-reinforced aluminium laminate (ARALL). Later, glass fbre-reinforced aluminium laminate (GLARE) was developed in which the glass fbre prepreg was used to supersede aramid fbre in FMLs to improve the overall performance. An attempt had been made to incorporate carbon fbre in the FMLs, but it was realised that the combination of aluminium alloys and carbon fbre results in galvanic corrosion. Galvanic corrosion is an electrochemical process happening when two dissimilar conductive materials are exposed to an electrolyte. In FMLs, the carbon fbre and aluminium can be electrically connected through the free edges of the laminates in the presence of an electrolyte, having a high risk of galvanic corrosion (Hamill et al., [2018](#page-15-0)). In this context, glass fbre is non-conductive, and thus the combination of glass fbre and aluminium will not result in galvanic corrosion. Since glass fbre-reinforced composites are not vulnerable to galvanic corrosion, they may serve as an insulating layer between carbon fbre-reinforced composites and aluminium layers (Ireland et al., [2012\)](#page-16-1). Overall, GLARE has been considered the most successful FMLs throughout the years. GLARE is mainly applied for the primary aircraft structures such as fuselage and wing skin (Alderliesten & Homan, [2006\)](#page-14-1).

The impact damages to the structures, which are due to the runway debris and hail strikes, are always the main concerns in aerospace industries. The impact velocity in some cases can be up to 100 m/s. Thus, GLARE has specifcally been optimised to improve its impact resistance. To date, the impact resistance of GLARE with various lay-ups has been extensively studied. Over the years, it had been concluded that GLARE could resist impact load more effectively compared to monolithic aluminium alloys. Abdullah et al. (2015) (2015) stated that FMLs based on glass fbre-reinforced epoxy exhibit superior impact resistance over aluminium alloys and carbon fbre-based laminates. Furthermore, GLARE also performs better than monolithic aluminium in resisting the fatigue load due to its sandwich confguration. Truthfully, the excellent fatigue performance of FMLs is mainly attributed to the fbre bridging mechanism where part of the load is transferred to the fbre, reducing the stress intensity factor and therefore lowering the fatigue crack propagation rate.

Despite the conventional FMLs showing outstanding characteristics, the long processing time and the high production cost have limited their use in a wide variety of structural applications. Generally, conventional FMLs are prepared through the autoclave process, which provides uniform structure and limited or zero void content in FMLs. Minimising the void content ensures the optimum mechanical performance of FMLs as the structural components in the aerospace sectors. Nevertheless, the autoclave process requires high cost due to the long processing time and tool preparation (Ramaswamy Setty et al., [2011](#page-17-0)). Therefore, alternative manufacturing process such as vacuum-assisted resin transfer moulding (VARTM) has been developed. This process requires a lower production cost than autoclave, but the quality of the product could not be achieved as high as the autoclave process. This issue remains a challenge among the research communities to develop a manufacturing method that could duplicate the performance of the autoclave process at a lower production cost.

The continuous rise of environmental awareness and consciousness has ignited an effort to implement cellulosic plant fbres in fbre-reinforced composites and FMLs. Plant fbres consist of cellulosic and non-cellulosic chemical contents such as lignin, hemicellulose, pectin, wax and impurities (Omran et al., [2021](#page-16-2)). It is wellknown that plant fbres have demonstrated some attractive properties such as low density, biodegradability, low energy consumption and high specifc properties. The high specifc properties of plant fbres are especially promising for improving the energy efficiency of the aircraft without compromising the mechanical performance. Nonetheless, plant fbres also possess some demerits which retard the use of natural fbres in primary structural applications. Quality variation, low thermal stability, poor compatibility with polymer matrices and high moisture uptake are the known drawbacks of plant fbres (Alsubari et al., [2021;](#page-14-3) Ilyas et al., [2021](#page-15-1); Nurazzi et al., [2021;](#page-16-3) Suriani et al., [2021a](#page-17-1)). Moreover, the plant fbres also demonstrate signifcantly lower mechanical strength than synthetic fbres. Among the approaches to improve the mechanical strength and the interfacial adhesion of plant fbre-based composites are the addition of nanofllers and the implementation of chemical treatments. The addition of fllers such as carbon nanotubes improves the mechanical performance of polymeric materials for several structural applications (Mohd Nurazzi et al., [2021](#page-16-4)). Various treatment methods have been established to enhance the interfacial bonding and reduce the moisture uptake of plant fbres. Apart from the addition of nanofllers and chemical treatments, the hybridisation concept has also been proposed to develop materials having balance in the mechanical properties and environmental friendliness. It is believed that the hybridisation concept could reduce the stress on the aerospace industries to search for lightweight, high damage tolerance and high toughness materials with the consideration of environmental friendliness. The current focus has been shifted towards the use of thermoplastic polymers and natural fbres.

2 Hybridisation Concept

Composite materials are composed of two or more phases with a boundary between components (Suriani et al., [2021b\)](#page-17-2). They have shown various outstanding benefts for structural components (Ng et al., [2020\)](#page-16-5). Hybridisation is the advancement in the feld of composite materials with the aim of compensating the disadvantages of one fbre with the advantages of the other fbre. The hybridisation concept is the current focus in the composite feld due to its several favourable characteristics. It can be defned as the incorporation of multiple types of fbres within the single matrix, allowing greater freedom to obtain the desired properties according to certain engineering applications. Hybridisation may include both natural and synthetic fbres in composite laminates and FMLs. The incorporation of natural and synthetic fbres imparts their respective advantages to the resulting hybrid composite materials,

achieving the balance in the eco-friendliness and acceptable mechanical properties. In addition to the fbre-matrix compatibility, fbre size, fbre orientation and the compatibility between multiple types of reinforcements also have a decisive effect on the mechanical properties of composite materials.

In hybrid composites, the hybridisation effect is more signifcant when the multiple types of fbres are strain compatible. Generally, the mechanical properties of hybrid composites are intermediate between their respective non-hybrid composites. Arpitha et al. ([2017\)](#page-14-4) reported that the mechanical properties of sisal/glass fbrereinforced hybrid composites were in between their respective non-hybrid sisal and glass fbre-reinforced composites. Sanjay and Yogesha [\(2018](#page-17-3)) revealed similar fndings where the mechanical properties of jute/kenaf/glass fbre-reinforced hybrid composites were intermediate between the non-hybrid composites. The incorporation of high-strength fbre in the composites tends to improve the mechanical properties. In certain cases, the mechanical properties of hybrid composites could be greater than non-hybrid composites (Sapuan et al., [2020;](#page-17-4) Ramlee et al., [2019;](#page-17-5) Rafquzzaman et al., [2016\)](#page-16-6). Hence, it could be foreseen that hybrid composites could have great potential to be used for structural applications in aerospace industries.

3 Standardised FMLs in Aerospace Sectors

Contemporary metallic alloys, particularly aluminium alloys, have been widely utilised in aerospace sectors over the past decades. Aluminium alloys have shown their high strength-to-weight ratio characteristic, and thus, they have gained popularity for a wide variety of structural applications. FMLs have been part of the third evolution materials in aerospace industries, which are developed to tackle the poor fatigue resistance of monolithic metals. However, it was also found that FMLs display excellent impact properties. Thus, the combination of metallic alloys and composite materials can resolve the poor fatigue performance of metallic alloys and also weak impact properties of composite materials. ARALL has been the frst generation of FMLs consisting of aramid/epoxy prepreg and aluminium alloys. In order to further improve the mechanical properties of FMLs, glass fbre was incorporated in the FMLs to replace aramid fbre. Besides ARALL and GLARE, carbon fbre-reinforced aluminium laminate (CARALL) was also developed due to the high stiffness characteristic of carbon fbre. Nevertheless, the consolidation of aluminium alloys and carbon fbre leads to galvanic corrosion, which deteriorates the mechanical properties of CARALL. Therefore, carbon fbre is generally isolated from aluminium alloys to avoid galvanic corrosion. In comparison with GLARE and ARALL, CARALL is still considered a novice, which is not widely used in industries. Figure [9.1](#page-4-0) shows the classifcation of standardised FMLs in aerospace sectors.

The standardised FMLs can be further classifed into different grades, which are based on their fbre orientations, the number of fbre layers and types of aluminium. As can be seen in Fig. [9.1](#page-4-0), GLARE and ARALL are sub-divided into six and four

Fig. 9.1 Classifcation of standardised FMLs. (Reprinted with permission from Feng & Malingam, [2019](#page-15-4))

grades, respectively. The majority of GLARE consist of aluminium 2024-T3 with an average thickness of 0.2–0.5 mm. However, the fbre orientation and the number of fbre layers may vary in each grade of GLARE. On the contrary, the fbre orientation and the number of fbre layers remain the same in ARALL. Each grade of ARALL has different types of aluminium alloy. For example, ARALL 1 consists of aramid prepreg bonded with aluminium 7075-T6; however, aluminium 2024-T3 is employed for ARALL 2. Apart from the FMLs based on aluminium alloys, titanium and magnesium alloys have also been used to form FMLs. Unlike aluminium-based FMLs which are widely used in aerospace sectors, titanium-based FMLs are primarily used in shipbuilding, military applications and off-shore industries because of their excellent corrosion resistance (Gorynin, [1999\)](#page-15-2). When exposed to oxygen and air, the oxide layer formed on the titanium alloys acts as a protective layer against corrosion. This oxide layer on titanium surface is even more durable than that of aluminium alloys (Golaz et al., [2013\)](#page-15-3). It should be emphasised that titanium alloys and carbon fbre have well electro-compatibility, and thus the galvanic-related issues could be avoided. Due to the superior fatigue and impact properties of FMLs, they have been proposed to be used in the automotive industries. In order to widen the application of FMLs in other sectors, the production cost of FMLs needs to be greatly reduced so as to gain acceptance by the industries.

In fact, these standardised FMLs have common disadvantages such as long processing time, poor inter-laminar fracture toughness and diffculty in repairing. Thus, an attempt has been made to replace the thermoset polymer with thermoplastic polymer in FMLs. Thermoplastic-based FMLs offer several benefts, including short processing time, ease of forming into complex shapes and excellent interlaminar fracture toughness. Numerous research studies have been conducted on the mechanical properties of thermoplastic-based FMLs (Feng et al., [2020;](#page-15-5) Subramaniam et al., [2019;](#page-17-6) Ng et al., [2017;](#page-16-7) Cortes & Cantwell, [2007;](#page-14-5) Reyes & Kang, [2007\)](#page-17-7). In this context, polypropylene (PP) is one of the most attractive thermoplastic polymers used in both composite and FMLs. PP has been commonly used as the polymer matrix for FMLs based on glass and carbon fbre-reinforced composites in aerospace and automotive industries (Gresham et al., [2006](#page-15-6); Mosse et al., [2006\)](#page-16-8). The low-density and low-cost characteristics make PP polymer extensively utilised as matrix material in composites and FMLs.

4 The Manufacturing Process of FMLs

The manufacturing process of FMLs comprises three major steps, including the metal surface treatment, metal-composite consolidation and post-stretching. Each of the steps plays an essential role in developing FMLs at their optimum mechanical performance. Metal surface pre-treatment is regarded as a critical step that cannot be neglected before the metal-composite consolidation. This pre-treatment aims at improving the metal-composite adhesion through surface modifcation. Several techniques have been utilised in aerospace industries to improve the wettability of the metal surface, consequently improving the adhesion quality between the metal and the composites. Mechanical, chemical, electrochemical, coupling agent and dry surface treatment are those typical techniques used in aerospace industries.

As mentioned earlier, the autoclave process is often the technique being used to manufacture the standardised FMLs for primary structural applications in the aerospace sectors. FMLs are formed by stacking the thin metal sheets with composite prepreg alternatively and then cured at a temperature of 120 °C and pressure of up to 6 bar (Botelho et al., [2006\)](#page-14-6). The purpose of maintaining high temperature and pressure during the autoclave process is to eliminate the void content in FMLs, allowing the quality of FMLs to meet the stringent standards established by the manufacturers. Void content is one of the critical elements that need to be controlled and minimised since the formation of the voids at the metal-composite interfaces results in delamination. At present, the autoclave process is the only production method for FMLs, which guarantees the high quality of the materials for the aerospace industries (Dariushi et al., [2019\)](#page-14-7). However, the high processing cost of the autoclave technique has limited the use of FMLs to other engineering applications.

In addition to the high processing cost, it was found that the exposure of FMLs to the elevated temperatures and thermal cyclic load in the autoclave undermines their mechanical properties (Müller et al., [2016](#page-16-9); da Costa et al., [2012\)](#page-14-8). Therefore, several manufacturing techniques, which are more economical and affordable, have been proposed, including VARTM, resin infusion technique and resin transfer moulding (RTM). Unfortunately, these out-of-autoclave techniques are not able to replicate the high performance of the autoclave process. The inability of out-of-autoclave techniques to control the high temperature and pressure entails non-uniformity of the laminates and formation of voids. The void content of FMLs manufactured using out-of-autoclave techniques is relatively high, which leads to the poor mechanical properties of the materials.

The consolidation of FMLs is followed by post-stretching to purge the residual stress in the FMLs. The thermal expansion coeffcients of the metal and composite layers are different, resulting in the unfavourable residual stress in the metal and composite layers. This residual stress adversely affects the fatigue resistance of FMLs. The increased crack opening and stress concentration factor resulting from the residual stress deteriorate the fatigue resistance of FMLs (Khan et al., [2009\)](#page-16-10). Accordingly, post-stretching is vital to reverse the residual stress in FMLs, thereby recovering the optimum fatigue resistance of the FMLs. The post-stretching mechanism of FMLs is demonstrated in Fig. [9.2](#page-6-0). During the post-stretching of FMLs, the metal layers are stretched into plastic deformation at a small percentage, whereas the fbre layer remains in the elastic region. The post-stretching converts the residual tensile stress in the aluminium to compressive stress. It had been shown that the

post-stretching process is indeed benefcial to the fatigue properties of FMLs. Lin et al. [\(1991](#page-16-11)) reported that GLARE and CARALL exhibited outstanding fatigue crack resistance after being subjected to the post-stretching process. The last step in the manufacturing process of FMLs is the inspection of any defects which could result in a detrimental effect on the mechanical properties.

5 Surface Pre-treatment of FMLs

FMLs encompass superior mechanical properties only when the metal-composite adhesion has reached an acceptable level. The metal-composite adhesion should be strong enough to allow the fbre bridging effect to take place in FMLs. In this regard, surface pre-treatment is the key factor that has a decisive effect on the mechanical properties of FMLs. The treatment modifes the wettability, surface morphology and surface energy of the metal sheets, thereby promoting the strong bonding between the metal and composite layers (Sinmazçelik et al., [2011;](#page-17-8) Kim et al., [2003\)](#page-16-12). As mentioned earlier, several types of surface pre-treatment methods can be applied to the metal layers. Generally, the combination of several surface pre-treatment methods is commonly practised in aerospace industries to guarantee the superior metal-composite adhesion. Figure [9.3](#page-7-0) shows the classifcation of metal surface pretreatments for adhesive bonding.

Fig. 9.3 Classifcation of the surface pre-treatments for adhesive bonding

5.1 Mechanical Surface Treatment

Among the surface treatment techniques, mechanical surface treatment is one of the most widely applied techniques owing to its simplicity (Mandracci et al., [2016\)](#page-16-13). In mechanical treatment, the process of roughening metal sheets is performed using abrasive paper, grit blasting or shot peening technology. The mechanical surface treatment produces a macro-roughened surface and eliminates the contaminants on the metal surface. This treatment results in physico-chemical and surface energy changes. Consequently, the wettability of the metal surface is greatly improved. In addition to abrasive paper, grit blasting is another advanced mechanical surface treatment technique that ensures the metal surface is free from contaminants and increases the surface roughness for a frm adhesion between metal sheets and composites. Grit blasting is conducted by spraying a continuous stream of abrasive particles such as sand or alumina to the metal surface to create a rough surface. Figure [9.4](#page-8-0) shows the three-dimensional (3-D) profle of steel surface before and after grit blasting. It can be observed that the surface roughness of steel after grit blasting, as shown in Fig. $9.4(a)$, is apparently higher than non-treated steel, as shown in Fig. [9.4\(b\)](#page-8-0). The selection of the types of abrasive particles highly depends on the substrate materials. Generally, the abrasive particles used in grit blasting should be harder than the substrate materials for abrasion purpose. Bresson et al. [\(2012](#page-14-9)) stated that it is highly recommended that the surface of metal sheets should be subjected to grit blasting prior to the adhesive bonding. Ng et al. ([2019\)](#page-16-14) concluded that the mechanical surface-treated aluminium laminates had higher lap shear strength than untreated aluminium laminates. Degreasing using a solvent such as acetone is usually carried out before the adhesive bonding to eliminate the contaminants or residues, which could hamper the development of chemical bonds.

Fig. 9.4 3-D surface profle. (**a**) Grit blasting. (**b**) Non-treated. (Reprinted with permission from Santhanakrishnan Balakrishnan et al., [2019](#page-17-9))

5.2 Chemical Treatment

Although mechanical surface treatment can improve the surface roughness of metal sheets, it may not guarantee the excellent metal-composite adhesion level as achieved by chemical treatment. Gonzalez-Canche et al. ([2018](#page-15-7)) evaluated the interfacial adhesion of FMLs using different surface treatments techniques, including degreasing, mechanical surface treatment and chemical treatment. They concluded that the metal sheets subjected to the chemical treatment had the highest wettability and surface roughness compared to degreasing and mechanical surface treatment. As a result, metal-composite interfaces had the highest shear strength after being subjected to chemical treatments. Moreover, chemically treated FMLs possessed the highest tensile properties in comparison with the degreased and mechanical surface-treated FMLs. Among chemical surface treatments, chromicsulphuric acid etching is the most widely applied chemical for metal surface treatment (Critchlow & Brewis, [1996\)](#page-14-10). Chromic-sulphuric acid etching involves the immersion of the metal into the solution consisting of sulphuric acid and potassium dichromate.

Chemical treatments often include both acid and alkaline treatments to obtain the desired results. Aluminium is a passive metal that has been widely used to combine with composite materials to form FMLs. This metal tends to react with oxygen and moisture content in the atmosphere to form aluminium oxide $(A₁,O₃)$ at the aluminium surface. It is essential to remove the weak oxide layer on the metal surface and substitute it with a new and stable oxide layer. Alkaline etching using sodium hydroxide (NaOH) has been a recognised oxide removing technique for aluminium alloys (Hu et al., [2019\)](#page-15-8). Besides removing the oxide layer on the aluminium surface, alkaline etching also modifes the surface topography, which eventually improves the metal-composite adhesion level. The reaction between aluminium alloys and sodium hydroxide (NaOH) can be represented by chemical Eqs. [\(9.1\)](#page-9-0) and [\(9.2\)](#page-9-1).

$$
Al_2O_3 + 2NaOH + 3H_2O \rightarrow 2Na[AI(OH)_4]
$$
 (9.1)

$$
2\text{Al} + 2\text{NaOH} + 6\text{H}_2\text{O} \rightarrow 2\text{Na} \left[\text{Al}\left(\text{OH}\right)_4\right] + 3\text{H}_2 \tag{9.2}
$$

As shown in Eqs. (9.1) and (9.2) , both aluminium and its oxide layer react with NaOH, leading to the formation of sodium aluminate. The subsequent chromicsulphuric acid etching eventually generates a new and stable oxide layer that can promote strong and durable adhesive bonds (Bishopp, [2005](#page-14-11)). Generally, the acid/ alkaline etching of metal alloys is followed by electrochemical treatment.

5.3 Electrochemical Treatment

It has been found that electrochemical treatment is able to improve the interfacial adhesion in FMLs signifcantly. Still, the performance of this treatment depends on various factors such as temperature, electrolyte concentration and voltage. Therefore, the temperature, electrolyte concentration and voltage should be carefully controlled to obtain the desired results. Xu et al. ([2016\)](#page-17-10) revealed that the optimal parameters in the anodising process could increase the surface energy of aluminium alloys, resulting in the maximum adhesion strength between aluminium and epoxy. Khan et al. [\(2017](#page-16-15)) performed a peel test on surface-treated CARALL. It was found that the electrochemical treatment further improved the peak force and fractured energy of CARALL when compared to those of CARALL with mechanical and chemical surface treatment only.

The alkaline/acid pre-treatment is often scarce for certain non-bonded area where corrosion may occur. Hence, electrochemical treatment is necessary to develop a durable protective layer with high wear and corrosion resistance on the metal surface before the coalescence of metal to composite layers. Anodising is often used in the electrochemical treatment, which gives a thin and ductile oxide layer to the metal layers. Chromic, oxalic, phosphoric and sulphuric acids are commonly used in the anodising process (Elabar et al., [2017](#page-15-9)). However, chromic-acid anodising (CAA) and phosphoric acid anodising (PAA) are the two most preferable electrochemical treatments in the aerospace industries. Through the anodising process on the metal surface, the corrosion susceptibility of the metal is greatly reduced. The reduction in corrosion susceptibility is attributed to the formation of a regular and uniform oxide layer with a high level of micro-roughness on the metal surface. The overall process of electrochemical treatment of aluminium can be expressed in Eqs. (9.3) and (9.4) (9.4) (9.4) (Wang et al., [2014;](#page-17-11) Vrublevsky et al., [2005](#page-17-12)). Due to the uniform pits formed during the anodising, polymer matrix can infltrate into the pits of the aluminium surface, providing a mechanical anchoring effect at the metal-composite interfaces. The mechanical anchoring effect endows the FMLs with excellent metalcomposite adhesion.

$$
2Al + 3H_2O \rightarrow Al_2O_3 + 6H^+ + 6e^-
$$
 (9.3)

$$
Al_2O_3 + 6H^+ \rightarrow 2Al^{3+} + 3H_2O \tag{9.4}
$$

5.4 Coupling Agent

Silane is regarded as the coupling agent which is commonly applied to improve metal-composite adhesion. Silane is a bi-functional molecule that could simultaneously react with the metal sheets and polymer matrices, forming strong chemical bonds with organic and inorganic layers. Indeed, silane can promote strong

Fig. 9.5 The bridging of silane coupling agent between inorganic and organic substances. (Reprinted with permission from Ebnesajjad, [2014](#page-15-11))

adhesion between inorganic metal layers and polymers by providing a bridging effect at the interface region. During the silane coupling agent treatment, the alkoxy groups of silane are hydrolysed to yield reactive silanol groups, and the aluminium sheets are soaked into the silane solution. The silanol groups form covalent bonds with the hydroxyl groups of aluminium sheets (Da Ponte et al., [2015\)](#page-14-12). Meanwhile, the other end of the silanol groups remains available to form covalent bonds with the polymers. The bridging of silane coupling agent between inorganic and organic substances is presented in Fig. [9.5](#page-11-0).

Apart from improving the adhesion at the metal-composite interface, silane coupling agent treatment can also reduce the rate of hydration of aluminium surface, and therefore, the metal-composite adhesion can be very stable in a humid environment. The penetration of moisture into the interfaces could weaken the adhesion and lead to physical detachment. Thus, the improvement in the resistance against hydrolysis resulting from the silane coupling agent is pivotal to ensure an excellent quality of the chemical bonding. Besides, the corrosion resistance of the metal-composite interfaces can be enhanced through the silane coupling agent treatment. Fedel et al. [\(2009](#page-15-10)) reported that the silane coupling agent could promote good adhesion between metal and polymer as well as provide a barrier effect against the oxygen and water. Sol-gel coating is also being used in aerospace applications to avoid corrosion which may lead to the delamination of FMLs. Furthermore, sol-gel coating for the aluminium sheets can prevent the occurrence of galvanic corrosion in CARALL.

5.5 Dry Surface Treatment

There are several types of dry surface treatments that have been developed for aluminium sheets. Excimer laser texturing, ion beam enhanced deposition (IBED) and plasma-sprayed coatings are grouped into dry surface treatment. Similar to the silane coupling agent, the dry surface treatment can reduce the use of toxic chromatebased treatments that negatively affects the environment. Excimer laser texturing has been shown to have a profound effect on bond strength and durability. This laser texturing alters the surface morphology and microstructure of the metal surface, leading to improved bond strength and durability. Laser texturing offers the fexibility to control the processing parameters. Hence, it is possible to have greater control on the surface topography of the metals at micro- or nano-scales (Dinca et al., [2015;](#page-15-12) Maressa et al., [2015](#page-16-16)). Galantucci et al. [\(1996](#page-15-13)) reported that the excimer laser texturing on the aluminium and carbon fbre-reinforced composites improved the bond strength.

IBED, also known as ion beam assisted deposition (IBAD), is a surface treatment that cleans and alters the surface by sputtering with high-energy argon ions in a vacuum environment (Sinmazçelik et al., [2011](#page-17-8)). This approach focuses on the nonequilibrium inter-diffusion and the formation of nucleation sites at the interfaces by low-energy ion bombardment to enhance the adhesion (Loh et al., [1987](#page-16-17)). Plasmasprayed coatings are also used as the surface pre-treatment method to improve the adhesion strength. Plasma-sprayed coating is a very versatile technique for the deposition of a wide range of materials, including metals, ceramic and composites. This process is performed by heating the materials, typically in powder, to the molten state, which is then projected towards the substrate at high velocity. The molten state of the materials and the high velocity impart excellent adhesion strength to the substrate (Davis et al., [1997](#page-14-13)).

6 Potential Applications of Hybrid FMLs

The contemporary FMLs have been involved in aerospace and marine applications. FMLs are primarily based on aluminium alloys and synthetic fbre-based composites in the aerospace industries. GLARE based on S-2 glass fbre/epoxy prepreg and aluminium alloys is currently being used for the primary aircraft structures such as the fuselage and wing skin materials. Similar to GLARE, ARALL fnds its application in aerospace industries for the wing skin panels and the cargo door (Alderliesten, [2009\)](#page-14-14). In contrast, CARALL is generally used as the impact absorber for helicopter struts and aircraft seats (Vlot & Gunnink, [2001](#page-17-13)). The most attractive feature of CARALL is the high stiffness of carbon fbre, which offers a superior fbre bridging effect to the laminates, resulting in a very low crack growth rate.

Considering the environmental friendliness of the materials without signifcantly compromising the mechanical properties, FMLs incorporated with both natural and synthetic fbres have shown their promising features in the replacement of synthetic fbre-based FMLs in aerospace industries. The use of natural fbre to supersede synthetic fbre is not realistic because the mechanical strength of natural fbres is lower than synthetic fbres. Thus, the hybridisation concept is considered an alternative way to be applied in both fbre-reinforced composites and FMLs. By properly designing the hybrid composites in a judicious way, the natural/synthetic fbre-reinforced hybrid composites could have comparable mechanical properties to nonhybrid synthetic fbre-based composites.

In recent years, intensive research studies have been conducted to explore the potential of hybrid composites to identify the feasibility of natural fbres in engineering applications. The previous literature studies concluded that hybrid composites and FMLs could have comparable mechanical properties to the non-hybrid synthetic fbre-based composites and FMLs when the high-strength synthetic fbres were located at the outermost layers in the laminates (Feng et al., [2019a,](#page-15-14) [b](#page-15-15)). This is because the outermost layers in the composite laminates are the primary loadbearing components (Ary Subagia et al., [2014;](#page-14-15) Idicula et al., [2010\)](#page-15-16). This phenomenon is even more signifcant when the composite laminates are subjected to out-of-plane loadings. Therefore, it is envisaged that FMLs based on natural/synthetic fbre-reinforced hybrid composites could be the excellent candidate to be applied in aerospace industries for the primary or secondary aircraft structures as substitute to synthetic fbre-based FMLs. By partially replacing the synthetic fbre in FMLs, it is believed that the energy efficiency of the aircraft will be increased as the density of natural fbres is lower than that of the glass fbre.

7 Conclusions

The advanced sandwich laminates, FMLs, have been successfully developed and applied for the primary structures in aerospace industries over the past decades. In addition to the excellent fatigue crack resistance of FMLs, these advanced sandwich laminates also lead to weight-saving and high impact resistance compared to conventional metal alloys. In order to obtain FMLs with optimum mechanical properties, the autoclave technique is usually used to manufacture FMLs with minimum or zero void content. However, this technique results in a high processing cost which hampers the use of FMLs to other structural applications in aerospace industries. Out-of-autoclave methods have been developed as an alternative, but these methods often result in a certain amount of void in the sandwich laminates. Thus, this issue remains a challenge to the engineers, scientists and researchers to develop a manufacturing method which can produce FMLs with high quality at low cost. Surface pre-treatment is a critical step to ensure excellent metal-composite adhesive bonding in FMLs. A strong metal-composite adhesion allows the effective fbre bridging mechanism, thereby improving the fatigue crack resistance.

Hybridisation has become the central focus for composite materials, aiming at reducing the dependence on synthetic fbre in both composites and FMLs. The previous studies have attested that hybrid composites may possess comparable mechanical properties to non-hybrid composites. This idea can be extended to the FMLs in the aerospace sectors in order to improve the environmental friendliness of the materials. Since natural fbres exhibit lower density than glass fbre, the energy effciency of air transportation could be enhanced as well. The replacement of nonhybrid synthetic fbre-based FMLs with hybrid FMLs could be achieved by proper material design. Therefore, further exploration of the natural/synthetic fbre-based hybrid FMLs is necessary to tackle the obstacles such as fbre-matrix compatibility and high moisture uptake when incorporating natural fbres in FMLs.

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