

# Review

## Reinforcement coatings and interfaces in aluminium metal matrix composites

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The interface between the matrix and reinforcement plays a crucial role in determining the properties of metal matrix composites (MMC). Surface treatments and coating of the reinforcement are some of the important techniques by which the interfacial properties can be improved. This review reports the state of art knowledge available on the surface treatments and coating work carried out on reinforcements such as carbon/graphite, silicon carbide (SiC) and alumina ( $\text{Al}_2\text{O}_3$ ) and their effects on the interface, structure and properties of aluminium alloy matrix composites.

The metallic coatings improved the wettability of reinforcement but at the same time changed the matrix alloy composition by alloying with the matrix. Ceramic coatings reduce the interfacial reaction by acting as a diffusion barrier between the reinforcement and the matrix. Multilayer coatings have multifunctions, such as wetting agent, diffusion barrier and releaser of thermal residual stress. The roles of reinforcement coating as a means of “*in situ* hybridising” and “*in situ* alloying” are described. © 1998 Kluwer Academic Publishers

### 1. Introduction

The need for new engineering materials with the advancement of modern technology in the areas of aerospace and automotive industries had led to a rapid development of metal matrix composites (MMCs). They can be tailored to have superior properties such as high specific strength and stiffness, increased wear resistance, enhanced high-temperature performance and better thermal and mechanical fatigue and creep resistance than those of monolithic materials. MMCs have an edge over polymer matrix composites because of their capability to withstand high temperatures, better transverse mechanical properties, superior thermal and electrical conductivities, excellent resistance to moisture, flame and radiation and zero out-gassing at vacuum.

Among the various matrix materials available, aluminium and its alloys are widely used in the fabrication of MMCs. This is because of the fact that they are light in weight, economically viable, amenable for production by various processing techniques and possess high strength and good corrosion resistance. Some of the important reinforcement materials used in the aluminium metal matrix composites are carbon/graphite, silicon carbide, alumina, zirconia and zircon in particulate, whisker or fibre form. Major fabrication methods used for aluminium metal matrix composites are stir casting, squeeze casting, compocasting, infiltration, spray deposition, direct melt oxidation process and powder metallurgy.

The interface between the matrix and the reinforcement is the critical region that is affected during the fabrication. If this interface is not tailored properly, it can lead to the degradation of the properties of the composites. The problems associated with the interfaces are the interfacial chemical reaction, degradation of the reinforcement, lack of wettability with the matrix, etc. These interfacial problems are system-specific. Hence, it is a difficult exercise to design optimized interfaces common and suitable for all systems. Some of the methods to obtain desired interfaces with better properties are the modification of the matrix composition, coating of the reinforcement, specific treatments to the reinforcement and control of process parameters. Among these, the most important technique to improve interfacial properties is that of coating of the reinforcement.

Reviews dealing with different aspects of interfacial problems and their effect on properties have been published earlier [1–9]. This paper aims to review the state of the art knowledge available on the surface treatments of reinforcements such as carbon/graphite, silicon carbide (SiC) and alumina ( $\text{Al}_2\text{O}_3$ ) and their effect on interface, structure and properties of aluminium alloy matrix composites.

### 2. Wetting

Wetting of a reinforcement by molten metal, an important aspect in MMC synthesis, is favoured by the

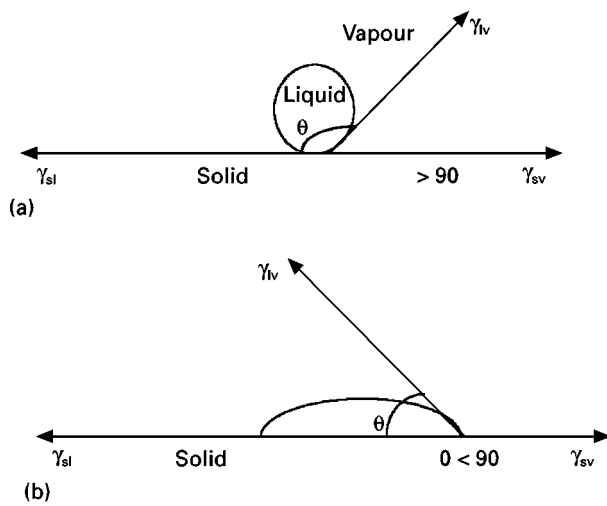


Figure 1 Schematic illustration of contact angle in a (a) non-wetting system, and (b) wetting system.

formation of strong chemical bonds at the interface. The presence of oxide films on the surface of molten metal and the adsorbed contaminant on the reinforcement surface generally leads to non-wetting of the reinforcement with molten metal. The wettability of a solid by a liquid is indicated by the contact angle, as shown in Fig. 1. The contact angle,  $\theta$ , between solid, liquid and gas/vapour is related by the Young–Dupre’s equation,

$$\gamma_{lv} \cos \theta = \gamma_{sv} - \gamma_{sl} \quad (1)$$

where,  $\gamma_{lv}$  is the surface tension of the liquid metal,  $\gamma_{sv}$  is the surface energy of the solid, and  $\gamma_{sl}$  is the solid/liquid interfacial energy. Based on the above equation, the contact angle,  $\theta$ , can be decreased by increasing the surface energy of the solid,  $\gamma_{sv}$ , decreasing the solid/liquid interfacial energy,  $\gamma_{sl}$ , or by decreasing the surface tension of the liquid,  $\gamma_{lv}$ . The liquid is said to wet the solid when  $\theta < 90^\circ$ , that is, when  $\gamma_{sv} > \gamma_{sl}$ . Some of the techniques to improve metal-reinforcement wettability include metallic coatings on the reinforcements, addition of reactive elements, such as magnesium, calcium or titanium, to the melt and heat treatment of particles before addition.

### 3. Nature of interface

The nature of interface has a strong influence over the properties of the metal matrix composites. Strengthening in the composites by the reinforcements is dependent on the strength of the interfacial bond between the matrix and the reinforcement. A strong interfacial bonding permits transfer and distribution of the load from the matrix to the reinforcement. The properties such as stiffness, fracture toughness, fatigue, coefficient of thermal expansion, thermal conductivity and creep are also affected by the nature of the interface.

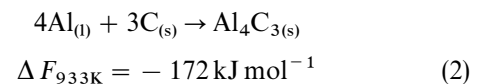
A mechanical bonding arises from mechanical interlocking between the matrix and reinforcements in the absence of all chemical sources of bonding and it is significant only in the case of fibre-reinforced com-

posites such as the brass–tungsten fibre system [10]. Chemical bonding occurs when the atoms of matrix and reinforcement are in direct contact and is accomplished by exchange of electrons. This type of bonding can be metallic, ionic or covalent. An interface with a metallic bond is more ductile than other bonds, and is desirable in metal matrix composites.

### 4. Interfacial chemical reaction

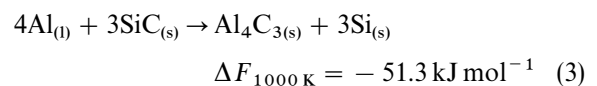
During processing of metal matrix composites, a chemical reaction occurs at the interface between the matrix and the reinforcement in some systems. In such cases, it leads to the formation of an interface reaction product layer with properties differing from those of either the matrix or the reinforcement. The extent of chemical reaction and the type of reaction products formed are dependent on the processing temperature, pressure and atmosphere, matrix composition and surface chemistry of reinforcements. Interfacial reaction can decrease the interfacial energy of the metal/reinforcement interface and improve adhesion through chemical bonding. The extent of the chemical reaction has a strong influence over the physical and mechanical properties of the composites. Further, the reaction products formed during processing may continue to form during service as well, thereby resulting in progressive improvement or degradation of the properties.

The following interfacial reaction is observed during the synthesis of carbon-reinforced aluminium metal matrix composites wherein the carbon can be either in the form of particulate or fibre based on graphite, pitch or PAN



The reaction tendency of carbon fibre with molten aluminium is observed to be severe when the melt temperature exceeds about 900 K. Aluminium carbide formation occurring by the degradation of fibres decreases their strength. A discontinuous reaction product has been observed at the interface of the aluminium–graphite particle system when the melt temperature and contact time exceed 1023 K and 4 h, respectively [11].

In silicon carbide-reinforced aluminium metal matrix composites, SiC is thermodynamically unstable in molten aluminium at around temperatures exceeding 1000 K [12]. The SiC reacts with molten aluminium [13, 14] to form  $Al_4C_3$ , rejecting metallic silicon according to the reaction



These reaction products have also been observed to cover  $SiC_p$  by Lee *et al.* [15] using SEM as shown in Fig. 2. However, the above reaction can be suppressed by having a matrix alloy containing a higher silicon content. Fig. 3 shows the silicon level required in the matrix to prevent the formation of aluminium carbide as a function of the melt temperature [16].

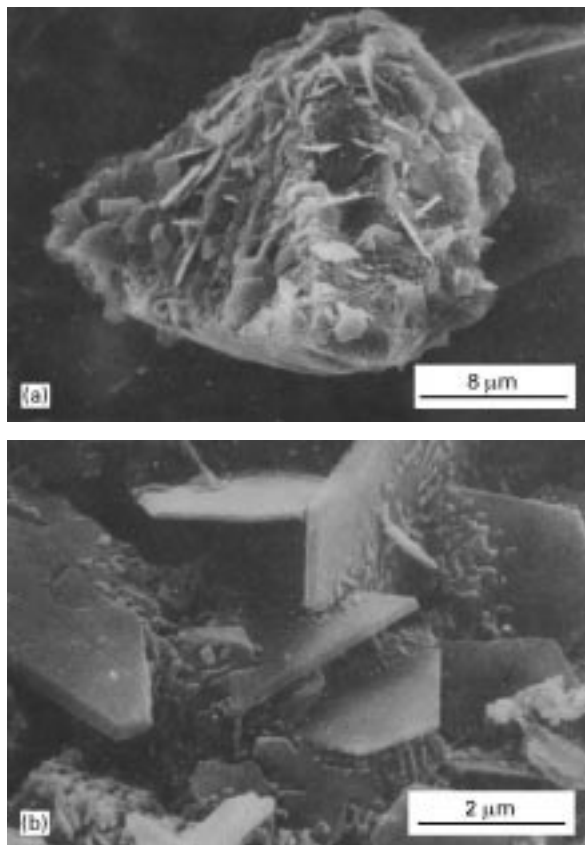


Figure 2 (a) Scanning electron micrograph of SiC<sub>p</sub> as covered with Al<sub>4</sub>C<sub>3</sub> and silicon crystals [15]. (b) Magnified views of Al<sub>4</sub>C<sub>3</sub> crystals having hexagonal platelet shape and dendritic shaped silicon crystals [15].

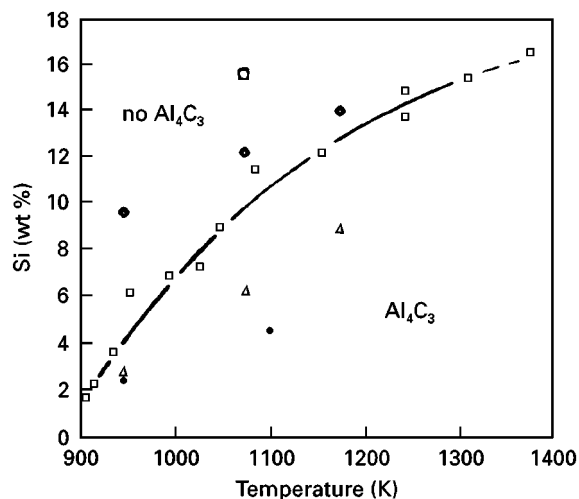
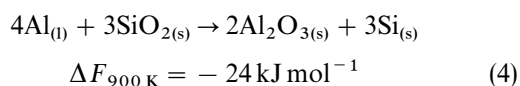
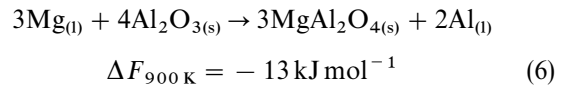
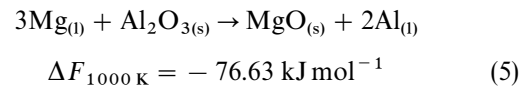


Figure 3 The silicon levels required in the matrix to prevent the formation of aluminium carbide as a function of melt temperature [16].

A few per cent of carbon and SiO<sub>2</sub> present in Nicalon fibres (β-SiC) react with molten aluminium to form Al<sub>4</sub>C<sub>3</sub> (Equation 2) and Al<sub>2</sub>O<sub>3</sub> + Si (Equation 4), respectively



Alumina (Al<sub>2</sub>O<sub>3</sub>) is considered as an ideal dispersoid because of its good interfacial compatibility and non-degrading surface with liquid aluminium. However, in most of the aluminium alloys of interest containing magnesium as an alloying element, magnesium reacts with alumina according to



MgO may form at high magnesium levels (> 1.5 wt % Mg) and low processing temperatures, while spinel forms at low magnesium levels (< 1.5 wt % Mg) [17]. Table I gives the possible reaction products and precipitates at the interface with various aluminium alloy matrices and reinforcement combinations [18–25].

## 5. Interfaces and composite properties

Most of the mechanical and physical properties of the metal matrix composites such as strength, stiffness, ductility, toughness, fatigue, creep, coefficient of thermal expansion, thermal conductivity and damping characteristics, are dependent on the interfacial behaviour.

The interface plays a crucial role in transferring the load efficiently from the matrix to the reinforcement. The strengthening and stiffening of composites are dependent on the load transfer across the interface. A high bond strength is required at the interface for effective load transfer. A strong bond is usually formed with the reaction between the matrix and the reinforcement, the reaction product determining the nature of the bond. A brittle reaction product at the interface makes the composite crack at lower strains. The presence of coarse intermetallic precipitates at the interface, as in Al–Cu–Mg/SiC composites, is also detrimental to mechanical properties. Even though the interface is free from reaction products, the tensile properties are dependent on the nature of the bonding. Studies on Al/SiC, Al/B<sub>4</sub>C, Al/TiC and Al/TiB<sub>2</sub> composites show that Al/TiC has the highest yield and ultimate tensile strengths [26]. This is due to the better bond integrity at the Al/TiC interface. In unidirectionally reinforced composites, the longitudinal tensile strength and crack growth initiation resistance are found to be insensitive to the nature of the interface. However, the weak interface exhibits extensive debonding and reduces the transverse and torsional strengths of the composite [27]. The ductility of the composites is also largely influenced by the interfaces [28, 29]. Al/TiC composites are more ductile compared to Al/Al<sub>2</sub>O<sub>3</sub> and Cu/Al<sub>2</sub>O<sub>3</sub> due to better interfacial bonding arising out of the metallic bond [28].

The toughness of the composite is influenced by crack deflection or fibre pull-out. In unidirectional composites, a weak interface is desirable for increased toughness when a crack propagates along the fibre, whereas a strong interface is required to prevent low-energy failure when a crack is parallel to the fibre. In

TABLE I Reaction products formed and precipitates at the interface with various reinforcements in aluminium alloys

Matrix	Reinforcement	Reaction products and precipitates	References
Al	C	Al <sub>4</sub> C <sub>3</sub>	[11]
Al	SiC	Al <sub>4</sub> C <sub>3</sub> , Si	[18, 19]
Al–Mg	SiC	Al <sub>4</sub> C <sub>3</sub> , MgO, Mg <sub>2</sub> Si, MgAl <sub>2</sub> O <sub>4</sub>	[20]
Al–Cu–Mg	SiC	CuMgAl <sub>2</sub> , MgO	[21, 22]
Al–Mg	Al <sub>2</sub> O <sub>3</sub>	MgAl <sub>2</sub> O <sub>4</sub>	[23]
Al–Cu	Al <sub>2</sub> O <sub>3</sub>	CuAl <sub>2</sub> O <sub>4</sub>	[24]
Al–Li	Al <sub>2</sub> O <sub>3</sub>	α-LiAlO <sub>2</sub> , LiAl <sub>5</sub> O <sub>8</sub> , Li <sub>2</sub> O	[24, 25]

particulate composites, when particles are more rigid than a matrix with a weak bond, the increased toughness is due to crack blunting effects and it appears that the same effect could be obtained by dispersion of voids [30]. When a bond is strong and the particles are less rigid than the matrix, an increase in toughness can be obtained by increasing the amount of material undergoing substantial massive plastic deformation. High residual stresses are developed in composites, when they are cooled from the processing temperature to room temperature due the mismatch in the coefficient of thermal expansion (CTE) between the fibre and matrix. As a result, within certain composite systems, radial, circumferential and/or longitudinal cracks are observed at the fibre–matrix interface region [31, 32]. In particulate composites, the strengthening is also attributed to the thermal mismatch strain present at the particle–matrix interface.

Fatigue properties of composites are also influenced by the interface. Investigations on Al/graphite composites show that the fatigue crack propagation rate (FCPR) is higher than in Al/zircon composites having better bond strength at the interface [33, 34]. This reveals that a weaker interface enhances the FCPR. Studies on the Al/SiC composite having a high bond strength at the interface show that a fatigue crack cannot propagate across the SiC particle unless it changes direction significantly and the crack deflection greatly reduces the FCPR [35].

Creep resistance of a particulate composite is determined with respect to creep threshold stress which is the index of resistance to creep. The creep threshold stress depends on the load transfer at the matrix–reinforcement interface which is dictated by the interface bond integrity. Al/TiB<sub>2</sub> *in situ* composites show higher creep resistance compared to powder metallurgy (P/M) processed Al/TiB<sub>2</sub> owing to the stronger interface and fine particle size in the former [26].

Studies on Al/15% TiC composites with 0.7 and 4 μm particles show that composites with 0.7 μm particles exhibit lower CTE due to a greater interfacial area. The lattice distortion is observed close to the matrix reinforcement interfaces. Studies on Al/SiC composites with particle sizes from 0.7–28 μm show that thermal conductivity increases with increase in particle size [36]. Fine particles have a larger interfacial area and the interface acts as a thermal barrier. 6090 Al/SiC composites with particles in the range 10–28 μm show a higher thermal conductivity than unreinforced alloy, probably due to an excellent bond-

ing at the interface [37]. Interfaces with reaction products act as stronger barrier to thermal conductivity than cleaner ones. In Ti/SiC composites with a 0.5 μm reaction layer of Ti<sub>5</sub>Si<sub>3</sub>, thermal conductivity is similar to that of unreinforced matrix. With a thick reactive layer (1 μm), the thermal conductivity reduces markedly.

## 6. Coating of reinforcement

Coating of a reinforcement is one of the successful techniques adopted to prevent the interfacial reaction and enhance the wetting of the reinforcement. Coating also prevents the diffusion of liquid metal into the reinforcement. Different types of coatings given to reinforcements are metallic, ceramic, bilayer and multilayer coatings containing metals and/or ceramics and are system-specific. The various coating techniques adopted aim at attaining a better, uniform and thin layer coating without degradation of the reinforcement properties. Some of the important coating techniques are chemical vapour deposition (CVD), physical vapour deposition (PVD), thermal spraying, sol–gel process, electrolytic, electroless and cementation methods. Because the type, purpose and process of the coating vary from system to system, they are discussed individually for the three most important reinforcements used in the aluminium matrix, namely carbon, silicon carbide and alumina.

### 6.1. Carbon

Carbon is one of the important reinforcements used in the fabrication of aluminium matrix composites. These composites find wide applications in aerospace, defence and electrical applications due to their high specific strength, high-temperature properties and excellent thermal and electrical conductivities. Carbon is most widely used in the form of graphite particles or fibres based on pitch or polyacrylonitrile (PAN). The major problems associated with the processing of carbon-reinforced aluminium metal matrix composites are the non-wetting of carbon by liquid aluminium at lower processing temperature (950–1050 K) and the reaction between carbon and aluminium at higher processing temperature. At around 1273 K [38], carbon becomes wetted and reacts with aluminium to form aluminium carbide at the interface which, in turn, reduces the strength of the composites [39, 40]. To overcome the above, various coatings are applied

to the carbon fibres to suppress the reaction and improve wettability at lower temperature. Techniques such as chemical vapour deposition, electrolytic and electroplating, are used to coat the fibres.

The important metallic coatings used to coat the fibres are copper [41–48] and nickel [45, 49–58]. Other coatings investigated include silver [59], vanadium [60], titanium [56, 61], molybdenum [56] and tantalum [56]. Nickel [62–66] and copper [67–70] coatings on graphite particles have also been studied. Nickel and copper coatings on carbon fibre prior to dispersion, enhanced its wettability with molten aluminium [71, 72]. The deposition of metals on carbon fibres is made by cementation, electroless and electrolytic processes. Among the various coating techniques mentioned above, the electroless coating method [42] has been reported to yield the best results with ultimate tensile properties of coated fibre near to that of uncoated fibres, whereas electrolytic and cementation processes exhibit lower tensile properties compared to uncoated fibres (Table II). However, coating thickness of 0.2–0.6  $\mu\text{m}$  yields better properties irrespective of the coating method adopted. With thickness below 0.2  $\mu\text{m}$ , coating is discontinuous and dendrite-type growth is observed above 0.6  $\mu\text{m}$  over the continuously coated fibre surface (Figs 4–6). The International Nickel Company (INCO) [51] is now commercially supplying nickel-coated carbon fibres and other refractory particulates for composite reinforcements with a very thin nickel coating (0.2–1  $\mu\text{m}$  range) through the Monds process. These observations have shown that apart from the coating material,

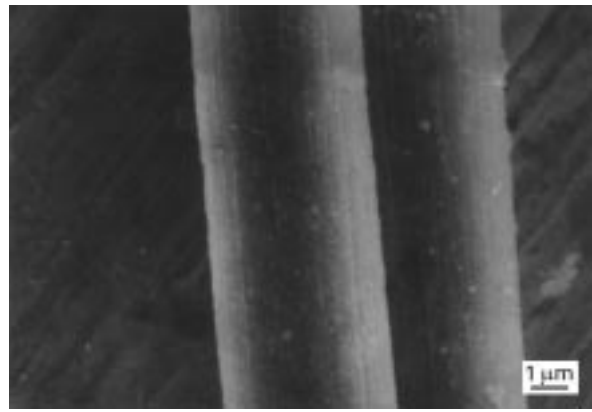


Figure 5 Completely coated carbon fibre; the thickness of the nickel coating is about 0.4  $\mu\text{m}$ .

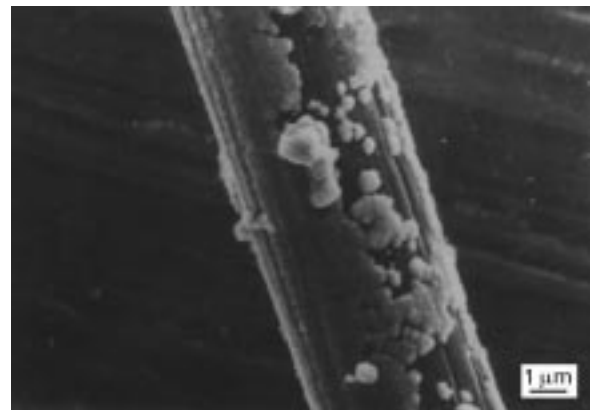


Figure 6 Fully coated carbon fibre; the thickness of the nickel is about 0.6  $\mu\text{m}$ . Dendritic-type precipitations are seen.

TABLE II Influence of metallic coatings and coating methods on the strength of carbon fibre [42]

Coating material	Coating method	UTS (GPa)	Elongation (%)
Uncoated		2.6–3.2	–
Ni	Cementation	1.5–1.81	0.8
	Electroless	2.4–2.81	1.23
Cu	Electrolytic	1.5–2.25	1.35
	Cementation	1.32–2.53	1.1
	Electroless	1.79–3.66	1.8

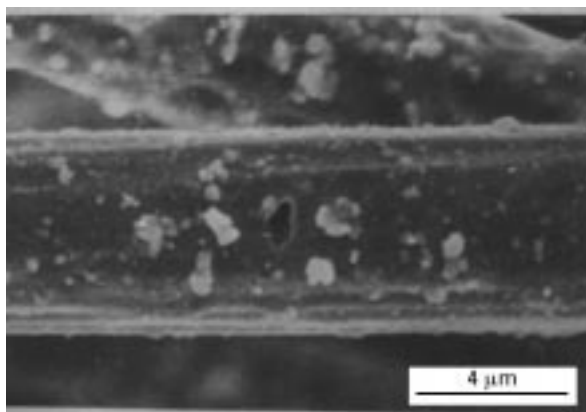


Figure 4 Partially coated carbon fibre; the thickness of the nickel coating is about 0.2  $\mu\text{m}$ .

coating techniques and thickness also play important roles in the properties of the fabricated metal matrix composites. The various metallic coatings given to carbon fibre and their influence on composites are described in Table III.

The nickel- and copper-coated fibres are found to change the matrix composition. For example, from calculations, a carbon fibre of 7  $\mu\text{m}$  diameter coated with 1  $\mu\text{m}$  thick nickel, when reinforced in aluminium to obtain a composite of 5 vol% carbon fibre, will add up around 16 weight per cent nickel to the melt. This leads to the formation of  $\text{NiAl}_3$  intermetallic compound which reduces ductility and increases the strength of the composites. Investigation of the cast Al–Si–copper-coated graphite composite [67] has shown lower strength and higher ductility than cast Al–Si–nickel-coated graphite composites. The presence of intermetallic compound improves the wear resistance of the composites. During fabrication, the coated copper dissolves into the aluminium matrix to form a solid solution, simultaneously exposing the surface to the melt directly, thereby improving the wettability of carbon by the aluminium matrix. The coating minimizes contamination of the dispersoid surface. The presence of  $\text{Al}_4\text{C}_3$  in isolated regions of the fibre–matrix interface in nickel-coated carbon-fibre dispersed composites (Fig. 7) and  $\text{CuAl}_2$  phases

TABLE III Important metallic coatings on carbon fibre and their effects

Coating material	Coating method and optimum thickness	Matrix	Composite fabrication technique	Effects	Reference
Copper	Electroless (0.3 $\mu\text{m}$ )	Al	Liquid metal infiltration	1. Improved wetting 2. Uniform distribution of fibres	[43]
Nickel	Electroless (0.2–0.4 $\mu\text{m}$ )	Al	Centrifugal pressure infiltration	1. Improved the throwing power of precipitant into the multifilament 2. $\text{NiAl}_3$ formation	[45]
Nickel	Electroless (0.2 $\mu\text{m}$ )	Al	Stir casting	1. Good wetting 2. $\text{NiAl}_3$ around fibres	[49]
Titanium	Chemical vapour deposition	Al	Liquid metal infiltration	1. Difficult to coat due to reaction 2. Rutile and anatase are formed	[61]
Silver	Electroless plating	Al-6063	Liquid metal infiltration	1. Promoted wetting 2. Interfacial reaction between Al and $\text{C}_f$	[59]

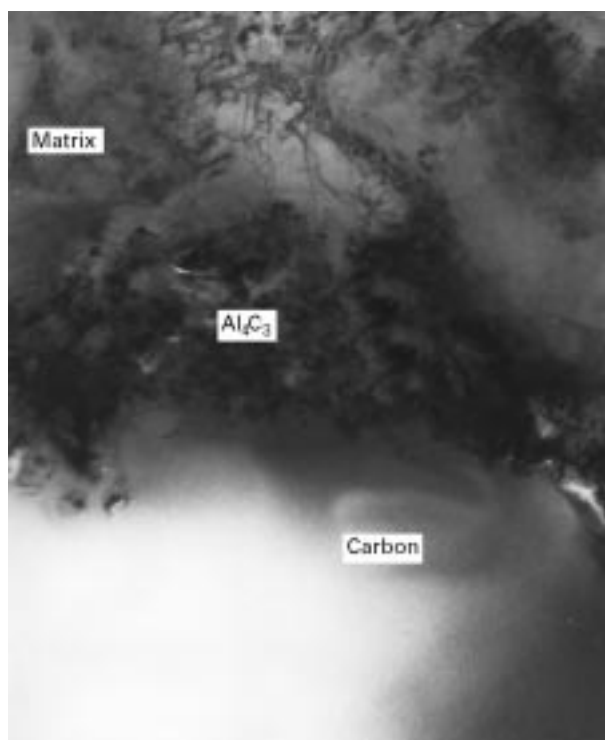


Figure 7 Interface between a carbon fibre and the matrix in electroless nickel-coated fibre dispersed composite showing  $\text{Al}_4\text{C}_3$  formation in some places (dark-field image).

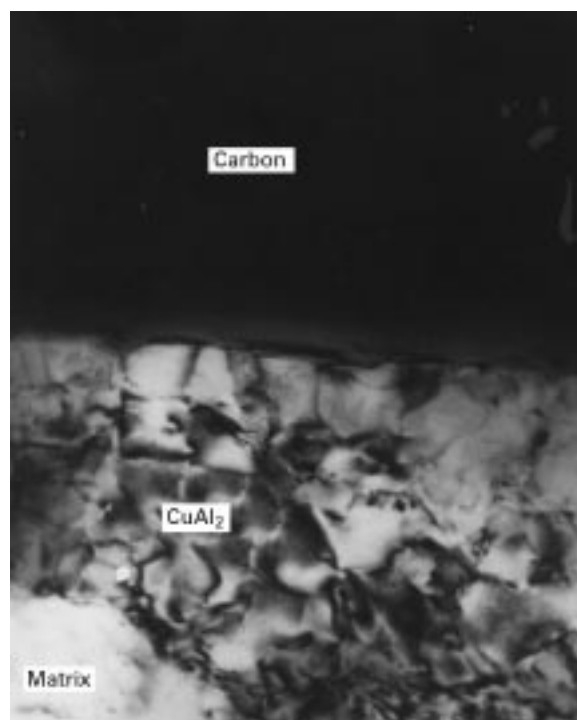


Figure 8 Interface between a carbon fibre and the matrix in electroless copper-coated fibre dispersed composite showing  $\text{CuAl}_2$  precipitate formation at the interface.

in copper-coated carbon-fibre dispersed composites (Fig. 8) have been observed. This has been attributed to the higher heats of formation of  $\text{NiAl}_3$  and  $\text{CuAl}_2$  phases. In the case of  $\text{NiAl}_3$ , the exothermic reaction increases its mobility, leading to its sweeping away from the interface exposing the fibre surface to molten aluminium [73, 74]. On the other hand, the endothermic nature of  $\text{CuAl}_2$  formation has helped in retaining it near the fibre–matrix interface, thereby keeping the fibres intact without undergoing reactions. This sug-

gests that selection of the coating material also plays an important role.

The ceramic coatings utilized include SiC [75–79], TiC [77],  $\text{B}_4\text{C}$  [77],  $\text{Al}_2\text{O}_3$  [80],  $\text{TiB}_x$  [81],  $\text{TiO}_2$  [82],  $\text{SiO}_2$  [58], ZrC [83, 84] and  $\text{TiB}_2$  [85]. The main function of ceramic coatings over carbon fibre is to act as a diffusion barrier to the aluminium matrix. The SiC has been coated using different techniques, such as chemical vapour deposition, solution coating, etc. Wang *et al.* [75] have developed a technique of

coating SiC on carbon fibre by benzene solution of polycarbosilane and reinforced SiC-coated fibre carbon in Al-10Si alloy. It has been observed that SiC coating effectively improved the resistance to oxidation of bare fibres and the wettability between the fibre and the molten metal. Thus, the interfacial chemical reaction is prevented (Fig. 9) thereby increasing the strength of composites to 189% of that with bare carbon fibres. B<sub>4</sub>C is effective in chemical protection

of the fibre as well as against oxidation during processing of the composites. TiC coating is not effective in the protection of fibres. The pyrolytic carbon coating on pitch P55 fibre improves the UTS in Al-4.5 Mg MMC from 576 MPa to 669 MPa. The effects of various ceramic coatings on interfaces of MMCs are given in Table IV.

The wetting of carbon fibres is also promoted by the presence of a reactive salt such as K<sub>2</sub>ZrF<sub>6</sub> [86,87]. Surface treatment of carbon fibres with an aqueous solution of K<sub>2</sub>ZrF<sub>6</sub> improves wetting by liquid aluminium at low processing temperatures. A decrease in contact angle from 165° to values as low as 50°–60° at temperatures slightly above the melting point of aluminium, has been observed. The reduction in contact angle is directly related to the amount of K<sub>2</sub>ZrF<sub>6</sub> spread on the fibre surface. The proposed mechanism for the wetting improvement involves two steps: (a) the reaction of K<sub>2</sub>ZrF<sub>6</sub> with aluminium to form K<sub>3</sub>AlF<sub>6</sub>, other fluoride species and intermetallics, and (b) dissolution of the alumina thin layer by the K<sub>3</sub>AlF<sub>6</sub> thus formed, enabling wetting of the carbon fibre.

In many cases, single-layer coatings have only limited functions, such as wetting agent or diffusion barrier. In addition to the above functions, coatings can have many other functions such as releasing the residual stress and adjusting the interfacial shear strength during fabrication, when suitable multilayer coatings are used. Some of the bilayer and multilayer coatings studied are pyrolytic carbon C<sub>p</sub>/TiC, C<sub>p</sub>/TiN, C<sub>p</sub>/SiC [88–90], SiO<sub>2</sub>/Cu [91], Al<sub>2</sub>O<sub>3</sub>/Al or Ni or Ti [80], C/Si [92] and C/SiC/Si [93,94].

The bilayer carbon–silicon gradient coating using chemical vapour deposition on the surface of carbon fibre has improved the oxidation resistance of the fibre greatly, and the tensile strength of C/Si coated fibre is higher than that of Si, SiC or SiO<sub>2</sub> monolayer-coated carbon fibre. The studies on C/TiC, C/TiN and C/SiC coated carbon fibres infiltrated with aluminium alloys have shown that C/SiC and C/TiN are more effective as a reaction barrier than C/TiC, and C/SiC double-layer coating is the best of these barrier coatings [87].

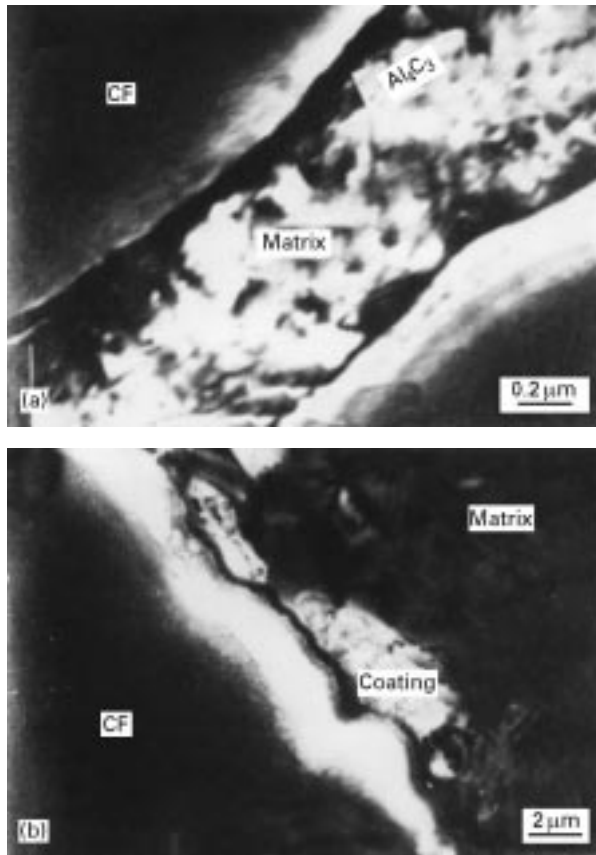


Figure 9 The interface of aluminium composites reinforced by (a) as-received carbon fibre showing Al<sub>4</sub>C<sub>3</sub> formation, and (b) SiC-coated carbon fibres [76].

TABLE IV Ceramic coatings on carbon fibres and their effects on interfaces in aluminium matrix composites

Coating material	Coating method	Composite fabrication technique	Effects	Reference
SiC	CVD	Squeeze casting	1. Effective protection of fibres during processing 2. Improved mechanical properties	[77]
Al <sub>2</sub> O <sub>3</sub>	Ion-plating	–	1. Good reaction barrier 2. Poor wettability	[80]
SiO <sub>2</sub>	SiO <sub>2</sub> coated and pre-treated with K <sub>2</sub> ZrF <sub>6</sub>	Gravity infiltration	1. Higher modulus of elasticity 2. Lower strength due to fibre degradation	[58]
TiO <sub>2</sub>	Sol-gel	Liquid infiltration	1. No reaction at TiO <sub>2</sub> /C interface 2. Improved wetting with formation of (Al, Ti)O <sub>2</sub> mixed oxide	[82]

To promote wetting of ceramic-coated carbon fibre,  $K_2ZrF_6$  treatment is a must which is evident from the full infiltration of SiC/ $K_2ZrF_6$ -coated carbon fibre in a short time with aluminium.

Alumina coating on PAN-based carbon fibre by ion-plating serves as an effective reaction barrier without much degradation in the fibre strength [79] but with poor wettability and bonding ability with aluminium. To enhance its wettability, aluminium, nickel or titanium has to be additionally coated to form a bilayer. Aluminium coating has good impact without degrading the strength of the fibre, whereas nickel and titanium coatings cause a remarkable degradation in the strength of fibre due to the formation of intermetallic compounds such as  $Al_3Ni$  and  $Al_3Ti$ .

A functionally gradient coating on carbon fibre for fabricating high-strength aluminium-carbon composites (UTS = 1250 MPa,  $V_f = 0.35$ ) has been achieved [92]. The multilayer consists of an inner pyrocarbon layer, an outer silicon layer and an intermediate gradient layer of C/SiC/Si and their optimum thicknesses are 0.1–0.15, 0.1 and 0.2  $\mu m$  respectively. It is possible to fabricate composites with carbon fibre coated with a pyrocarbon layer, exhibiting a UTS up to 1400 MPa but with very weak shear strength [93]. The multilayer C/SiC/C coating results in a good interfacial shear strength and also serves as a releaser of thermal residual stress, apart from providing improved wettability and a good diffusion and reaction barrier.

## 6.2. Silicon carbide

The aluminium-SiC composite system finds potential applications as structural elements in the automotive and aerospace industries. These composites possess unique properties such as improved strength, modulus and wear resistance and good resistance to corrosion. But several drawbacks of these materials such as low-temperature ductility and poor toughness, hinder their wide range of application. The causes for the remarkable drop in ductility and toughness of the composite are believed to be related to the structure at the interface region and the processing factors. The major problems encountered during the fabrication of SiC-reinforced aluminium matrix composites are the reactivity of SiC with molten aluminium at higher processing temperatures and the poor wettability of SiC at lower processing temperature (900–1000 K). The reaction between SiC and liquid aluminium during processing causes significant degradation in the properties of the composites [95–99]. In order to prevent the degradation of SiC (particles, whiskers or fibres) and improve wettability, various treatments and coatings have been attempted.

The metallic coatings given to SiC are copper [100–102], nickel [102–104], antimony [105] and silver [102]. Investigation by Moon and Lee [102] has shown that the wettability of copper-, nickel- and silver-coated SiC fibre with aluminium is better than as received fibre, copper and silver coatings being more effective. The driving force for wetting has been considered to be increased by the interfacial reaction

between metallic thin film and the liquid aluminium. When the interfacial reaction involved is a eutectic reaction, the interfacial free energy is sufficiently reduced to improve the wetting behaviour and the  $Al_2O_3$  film is easily broken down. It is also observed that the surface of the copper coating has only a few monolayers of CuO while the surface of the nickel coating is metallic nickel [104].

The influence of various ceramic coatings as a possible barrier against degradation of SiC particles with aluminium, has been understood. Some of the important ceramic coatings studied include  $SiO_2$  [106–114], BN [115],  $Al_2O_3$  [106, 109, 116–118],  $TiO_2$  [106, 119], MgO [120] and oxides of antimony and tin [121]. Other types of coatings investigated are Ni–P [122, 123], bilayer coatings of TiC/C [124] and coatings with reactive salts such as  $K_2ZrF_6$  [87].

Table V lists the various ceramic coatings given to silicon carbide and their effects on the interface and properties of aluminium composites. The oxide coating on SiC has good impact as a barrier coating by reducing the degradation of SiC during composite fabrication. The  $SiO_2$  coating is given mainly by the thermal oxidation of the SiC particulates [113]. During the fabrication of the composites based on Al–Si–Mg alloys with oxidized SiC particles, the  $SiO_2$  layer reacts with liquid aluminium alloy to form a polycrystalline layer of  $MgAl_2O_4$  spinel and  $Mg_2Si$ . In high magnesium content alloys, fine MgO crystals are more likely to form than  $MgAl_2O_4$ . Hence, in the as-cast condition,  $SiC_p$  are covered by complex oxide layers. Remelting studies of these composites carried out at 1073 K have revealed that the oxide layer is stable at this temperature and less degradation of the particles occurs compared to composites reinforced with as-received particles. The thickness of the  $MgAl_2O_4$  layer depends on the thickness of the  $SiO_2$  layer on the SiC particles. A lower oxidation level leads to a thinner layer of  $MgAl_2O_4$  in the composite, resulting in less-effective protection. However, a thicker layer results in a brittle interface, thus lowering the strength of the composite.

The other type of oxide coatings is mostly carried out by the sol-gel or dry-mixing technique. The SiC particles have been coated with  $Al_2O_3$  and MgO using the sol-gel technique [125]. The coating is about 50 nm thick. Fig. 10 shows the silicon content in the commercially pure aluminium as a function of exposure time of coated and uncoated  $SiC_p$  in the melt at 990 K. There is a significant reaction between the particles and liquid aluminium during infiltration at 990 K. The  $Al_2O_3$  and MgO coatings on  $SiC_p$  significantly reduce the rate of reaction, the latter being more effective than the former.  $TiO_2$  coating obtained by a dry mixing process has been found to be discontinuous on  $SiC_p$  as well as ineffective for durations longer than 2 h at higher temperatures during processing and remelting, leading to degradation of SiC with the formation of  $Al_4C_3$  crystal and silicon release. The above observation suggests that dissolution of  $TiO_2$  coating occurs when the  $TiO_2$ -coated  $SiC_p$  comes into contact with the liquid metal. BN coating is also given to SiC whiskers [115]. SiC whiskers are precoated



TABLE V Major ceramic coatings on SiC and their effects on the interface and properties of composites

Coating material	Coating method	Matrix	Composite fabrication method	Effects	Reference
SiO <sub>2</sub>	Dry mixing process	Al–1Mg	Stir casting	Significant reduction of Al <sub>4</sub> C <sub>3</sub> formation at 973 K and no protection above 1073 K	[106]
SiO <sub>2</sub>	Thermal oxidation	Al	Pressure infiltration	Once Al reacts with SiO <sub>2</sub> , reaction between Al and SiC proceeds	[107]
SiO <sub>2</sub>	Oxidation of SiC <sub>p</sub>	Al and Al–Mg (5083)	Compcasting	Interfacial reaction is dependent on alloy composition and thickness of SiO <sub>2</sub> layer	[108]
Al <sub>2</sub> O <sub>3</sub>	Dry mixing process	Al and Al–1Mg	Stir casting	Increase in interfacial reaction	[106]
Al <sub>2</sub> O <sub>3</sub>	Sol–gel	Al	Infiltration	Good protection in whiskers, but no protection in particulates	[116]
TiO <sub>2</sub>	Dry mixing process	Al and Al–1Mg	Stir casting	Remarkable reduction in Al <sub>4</sub> C <sub>3</sub> formation	[106]
SnO <sub>2</sub> and SbO <sub>2</sub>	Sol–gel	Al and Al–Mg	–	Not very effective	[121]
TiO <sub>2</sub>	Sol–gel	Al–1Mg	Stir casting	MgO/Ti reaction layer formed in the interface is responsible for the protection of particles	[119]

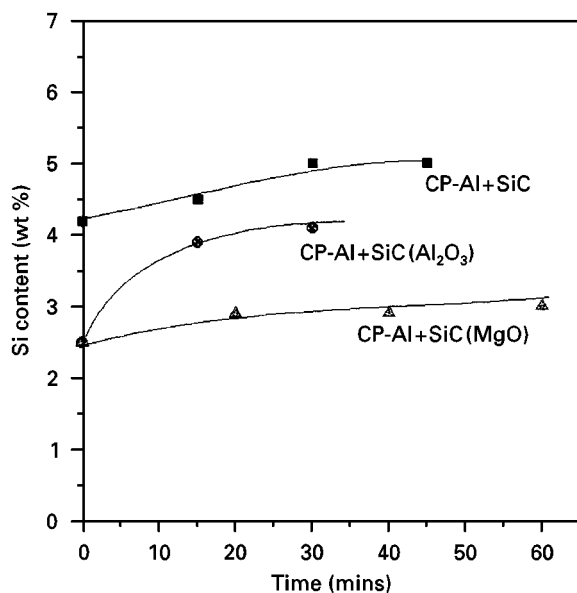


Figure 10 The silicon content in the commercially pure aluminium as a function of exposure time to the melt at 990 K of coated and uncoated SiC particles. The silicon content at time zero corresponds to that measured after fabrication of the composite (after [110], replotted).

with a polymer which is then thermally degraded to become highly porous carbon. This coating absorbs inexpensive borate solutions which are converted to a strongly adhering boron nitride layer at a high temperature in a nitrogen or ammonia atmosphere. Thickness can be predicted using a correlation based on the amount of polymer precoat used, the time and the amount of borate absorbed. TiC/C coating on SiC fibres by a closed-field unbalanced magnetron sputter

ion plating technique has been observed not only to improve the wettability of SiC but also protect the fibre against degradation when in contact with molten aluminium [124].

### 6.3. Alumina

The alumina-reinforced aluminium metal matrix composites find wide application next to carbon- and silicon carbide-reinforced composites in the areas of automotive and aerospace industries. Al–Al<sub>2</sub>O<sub>3</sub> metal matrix composites possess high elevated-temperature strength, wear resistance, damping properties, electrical conductivity, thermal conductivity and coefficient of thermal expansion. The alumina can be in the form of particulates, whiskers and fibres.

The alumina in a pure aluminium matrix is considered to be the ideal dispersoid with no chemical reactions. But, when aluminium alloys are used as the matrix, the Al<sub>2</sub>O<sub>3</sub> reacts with alloying elements such as magnesium. The other major problem is its lower wettability below 900 K [126]. In order to enhance its wettability, metallic coatings such as nickel [110, 127], cobalt [128, 129] and palladium [130] have been applied to alumina. MgO-coated alumina particles [131] have been found to improve the properties of composites compared to as-received ones. The deposition of nickel on alumina is made by nickel ions from a solution under hydrogen pressure in the presence of ammonia as a complexing agent. The optimum conditions devised for coating nickel include an  $[[\text{NH}_3]/\text{Ni}^{2+}]$  ratio of 1:6, a hydrogen pressure of 2760 kPa at 448 K, and an anthraquinone addition of 0.04 g l<sup>-1</sup> [126]. Cobalt coating using the sol–gel technique increases its wettability during processing. The

evaluation of tensile properties and fracture behaviour of cobalt-coated  $\text{Al}_2\text{O}_3$  fibre-reinforced 2024 Al alloy composites has shown improved properties compared to that with uncoated fibres. The formation of  $\text{Al}_9\text{Co}_2$  and  $\text{Co}_2\text{O}_3$  phases has been observed at the interfaces [128]. The fractographic analysis of the above composite has revealed a change-over of fracture behaviour from interfacial debonding to matrix deformation.

## 7. General discussion

This review describes the various coatings given to the reinforcements and their effects on the interfacial behaviour and properties of the aluminium metal matrix composites. In general, the coatings given to reinforcements can be classified into monolayer and multilayer coatings. Monolayer coatings include metallic and non-metallic coatings. The multilayer coatings can be classified into bilayer and multifunctional multilayer coatings. The reinforcement coatings can also be classified with respect to the function of the coating in the composites as wetting coating, barrier coating and multifunctional coating. The wetting coating promotes wetting of reinforcement with the matrix during processing and the barrier coating acts as a diffusion barrier between the matrix and reinforcement. The multifunctional coating may have more than one function as wetting promoter, barrier layer, releaser of residual thermal stresses, etc.

Among the various types of coating described above, the metallic coatings function as the wetting promoter of the ceramic reinforcement in liquid aluminium. They also protect the reinforcement from the reaction with the matrix, to some extent. The improvement in wetting is brought about either by the dissolution of the coated metal into the aluminium matrix or the formation of reaction products with aluminium or any one of the alloying elements present in the matrix alloy. Nickel-coated carbon or SiC reinforcement leads to the formation of brittle  $\text{NiAl}_3$  intermetallic phase, resulting in considerable decrease in ductility. However, improvement in Young's modulus and yield strength and a marginal increase in ultimate tensile strength have been observed. The fracture properties of the composites are mainly controlled by SiC particle fracture and the  $\text{NiAl}_3$  phase has only a mild influence. In the case of silver coating, the high solubility of silver in aluminium provides good wetting without forming any intermetallic phases. The coating process and the thickness of the coating also influence the fibre strength and hence the properties of composites. The electroless method is observed to give better tensile properties than electrolytic and cementation methods on carbon fibres. A continuous coating is obtained at 0.2–0.6  $\mu\text{m}$  thickness, above which dendritic growth on the surface of the fibre is observed. Development of nanometre thick stable and continuous coating requires serious attention.

The ceramic coating acting as diffusion barrier between the matrix and the reinforcement reduces the interfacial reaction. Oxide coatings such as  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$  and  $\text{TiO}_2$  on SiC give protection during fabrica-

tion of the composites. However,  $\text{Al}_4\text{C}_3$  formation is not fully prevented during processing at higher temperatures, i.e. above 1050 K or holding the melt for longer time. This is either due to the dissolution of ceramic coating into the melt or reaction of the coating with some elements present in the matrix. The magnesium in the matrix can react with the  $\text{Al}_2\text{O}_3$  to form the spinel. TiC, TiN and  $\text{B}_4\text{C}$  coatings give better protection to the reinforcement than oxide coating with satisfactory wetting. The right choice of coating method plays a critical role in fabricating better composites. This is because of the observation of discontinuous  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  coatings on reinforcements resulting from the dry-mixing or sol-gel processes.

The limited functions of monolayer coating lead to the development of multilayer coatings with multifunctions. The C/SiC/Si functionally gradient coating on carbon fibre results in successful production of C/Al composites with an ultimate tensile strength up to 1250 MPa [93]. Here, the functions of various layers include SiC as a diffusion layer, silicon as a wetting promoter, and soft pyrocarbon layer to reduce the stress created by stiff SiC coating which could cause fracture at lower stress. Even though multilayer coatings are costlier, most of the interfacial problems can be solved.

One major problem in the metal matrix composites is its lower toughness. The interfacial region which is less tough than the neighbouring bulk material, allows the crack to propagate. The lower toughness of the interface is mainly due to its nature of bonding. The covalent bonding arising from brittle interfacial products leads to lower toughness. It is necessary to find a suitable coating which provides a metallic type of bond to obtain higher toughness.

Apart from the above functions such as wetting promotion and diffusion barrier, the coatings can also act as “*in situ* hybridizing” and “*in situ* alloying” agents. This would make the coatings multifunctional and lead to cost-effective fabrication of metal matrix composites. A suitable metallic coating on the reinforcement can act as wetting promotion agent as well as an *in situ* hybridizing agent. An intermetallic phase, if formed between the coating and the matrix, would strengthen or improve the wear resistance of the composites. The dissolved coating in the matrix can form a second hybridized reinforcement by reacting with the coated material in the presence of a suitable non-metallic gas. This means of adding a second reinforcement into the matrix can be termed *in situ* hybridization of composites. Aluminium/intermetallic composites such as Al/ $\text{Al}_2\text{Cu}$  [132–134] and Al/ $\text{Al}_2\text{Ni}$  [134, 135] have been studied. Similarly, the intermetallics formed from copper and nickel coating can be distributed properly in the matrix to enhance the composites properties. It is shown that the presence of intermetallics improves the strength of the composites. When certain metal coatings dissolve in the matrix after serving their purpose of wetting the reinforcement by the matrix, they can enhance the properties of the matrix and hence the composite by acting as an alloying element. This method of alloying addition can be termed *in situ* alloying of composites. In this

case, judicious selection of an initial matrix alloy is necessary to end with the required matrix alloy composition during composite synthesis with coated reinforcements. Studies have been done with strontium coating on fibres preferably in the form of SrO. The coated strontium functions both as wetting promoter and modifier for eutectic silicon, when the fibres are infiltrated by Al-Si alloys [136]. Further work needs to be carried out on 'in situ hybridization' and 'in situ alloying' techniques during fabrication of composites and the key factor in this context is controlled coating thickness.

## 8. Conclusion

The application of coatings to the reinforcement during fabrication of metal matrix composites is an important step, having promising effects on the interfacial, physical and mechanical properties of the composites. There are different types of coatings, namely, metallic, ceramic, bilayer, multilayer, etc., which are system-specific in nature. Metallic coatings improve the wettability of the reinforcement and prevent the excessive interfacial reaction by enrichment of the coated metal in the matrix near to the interface. The metal coating process is economically viable, but leads to unwanted alloying. A ceramic coating reduces the interfacial reaction by acting as a diffusion barrier between the reinforcement and the matrix. Most of the ceramic coating techniques are expensive. Multilayer coating leads to the multifunctional behaviour of the coatings such as wetting agent, diffusion and reaction barriers and releaser of thermal residual stress. Reinforcement coating can act as 'in situ hybridizing' and 'in situ alloying' agents, during the fabrication of the composites. However, appropriate techniques need to be developed for achieving controlled thickness coatings. Both multifunctional coatings and the effect of coating as hybridizing and alloying agents need detailed study.

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