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ARTICLE

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A REVIEW ON MANUFACTURING OF AEROSPACE GRADE CARBON FIBER AND PREPREGS

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Aerospace-grade carbon fibers (CFs) are specifically engineered to qualify for aviation environments. CF's mechanical strength and durability come from the precursor material and depend on the processing method. Polyacrylonitrile (PAN)-based precursor is mainly used to make high tensile strength CF suitable for airframe structures. This article critically reviews different aspects of making precursors for CF, their processing, and fabrication of CF prepregs. Current trends and future projections of CF for aerospace sectors have also been included.

Keywords: *Aerospace-grade carbon fibers; precursor material; polyacrylonitrile; carbon fiber prepregs*

Introduction

Initially, the aerospace industry was sceptical with the idea of incorporating fiber-reinforced polymer (FRP) composites because of their low inter-laminar strength and poor durability under hot-wet (hygrothermal cycling) and other environmental conditions. However, the advent of commercial carbon fiber in the 1960s marked a turning point. The industry began to appreciate the potential of CF in airframes and engines thanks to its strength, lightweight, stiffness, and resistance to fatigue and corrosion. However, the commercial risks associated with using these composites for primary aircraft structures, which necessitated a complete overhaul of production facilities, led to the continued use of aluminium as the primary structural material despite its fatigue and corrosion issues [1].

The high cost of CF and limited understanding of design rules initially hindered the widespread use of CF composites, confining their application to semi-structural components. However, the OPEC energy crisis in the 1970s presented a unique opportunity. The crisis, coupled with the inherent corrosion problems of aluminium, led to a surge in using CF composites in fighter aircraft and commercial airlines. The cost of manufacturing CF also decreased, thanks to improved design and efficient process equipment, further fuelling their adoption in aircraft during the 1980s and 1990s [2].

The selection of materials for aerospace applications is a highly specialized process requiring a deep understanding of aerospace structures. The evolution of airframe materials is a testament to this. From the early days of wood and fabric, the aerospace industry has transitioned to modern carbon-fiber-reinforced polymer (CFRP) composites [3]. The primary selection criteria for airframe materials are the design, function, loads, and environmental service conditions. Safety-critical structures, such as the fuselage, wings, landing gear, and empennage, demand materials with high mechanical properties and excellent durability in the aviation environment. These materials must also be lightweight, as the airframe typically accounts for 20-40% of the all-up weight of most aircraft. Therefore, using structurally efficient but lightweight

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materials not only saves weight but also leads to less fuel burn, improved range and speed, and smaller engine requirements[4].

The Aircraft Energy Efficiency (ACEE) program taken by NASA (1975–1985) in collaboration with Boeing, McDonnell Douglas, and Lockheed was the first attempt to introduce CFs in commercial aircraft—the program aimed to design and fabricate CFRP parts for commercial aircraft use [5]. CFRP-based B737 spoilers, a B727 elevator, and a B737 horizontal stabilizer torque box were the few successes of the program. Among these, the B737 horizontal stabilizer torque box, still in commercial service, was Boing's first primary structure made of CFRP. A standard modulus grade CF with an untoughened epoxy resin cocured at 175°C with aramid paper honeycomb core was used to fabricate the B727 elevator, and a similar design concept was employed in making inboard ailerons, elevators, and rudders. PAN fiber-based standard modulus grade (T.M. 220 GPa) CF reinforcement for 120°C and 175°C cured epoxy matrices was used in making B737 spoilers and outboard ailerons[6].

An intermediate-modulus grade (T.M. 290 GPa) CF prepregs were used to fabricate B777 empennage and floor beams. All these initial successes convinced Boing to manufacture the entire fuselage, wing, and empennage skins and much of the support structure of the successor B787 aircraft from CFRP[6]. Please refer to Fig. 1 to note the progressive advancement. CFRP composite parts are lighter than aluminum counterparts, but their costs are significantly higher. Reducing part counts, however, can offset this disadvantage as many smaller composite parts can be bonded to make a more monolithic structure, which will be competitive with the cost of fabricating and assembling multiple parts to form a metallic structure. The present article reviews the manufacturing of carbon fiber and its prepregs for aerospace applications. Synthesis of precursor polymer and making of fiber through spinning are elaborated. Various critical stages of heat conversion from precursor to carbon fiber are also given. Resin formulations for making prepreg and its manufacturing aspects have been detailed. This article also gives the present status and future projections for using carbon fiber composites in aerospace structures.

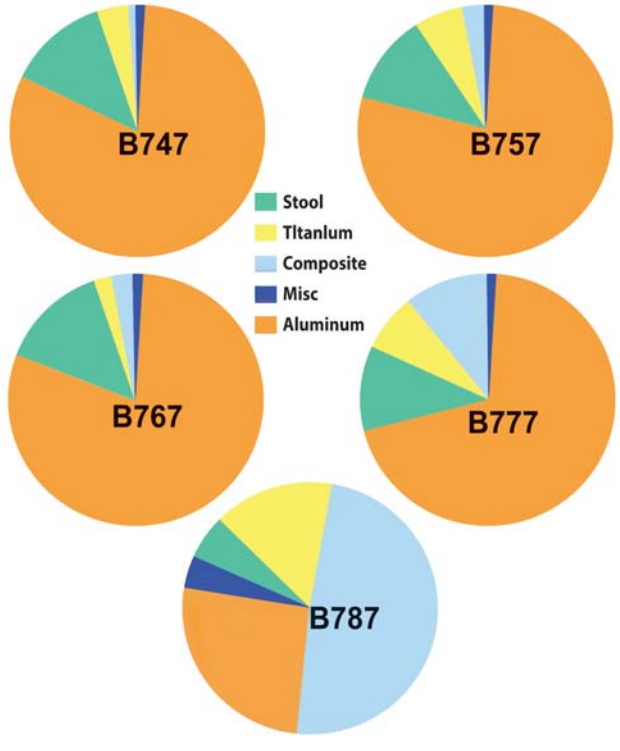

Fig 1: Distributions of structural materials used on selected Boeing [Reproduced after permission from Reference 6].

Precursor Polymer

Polyacrylonitrile (PAN), pitch, rayon, and cellulose are the most used precursors to manufacture different grades of CFs[7]. Among these, PAN contributes to ~90% of CF manufacturing owing to its structural advantages in producing high-strength CF. However, PAN homopolymer has disadvantages in subsequent processing stages, such as low solubility during spin dope preparation because of structural rigidity and uncontrolled heat generation because of high exothermic reaction during thermo-oxidative stabilization leading to decomposition, cross-linking, and even chances of fire[8]. These problems are not encountered with PAN copolymer as the comonomers hinder the

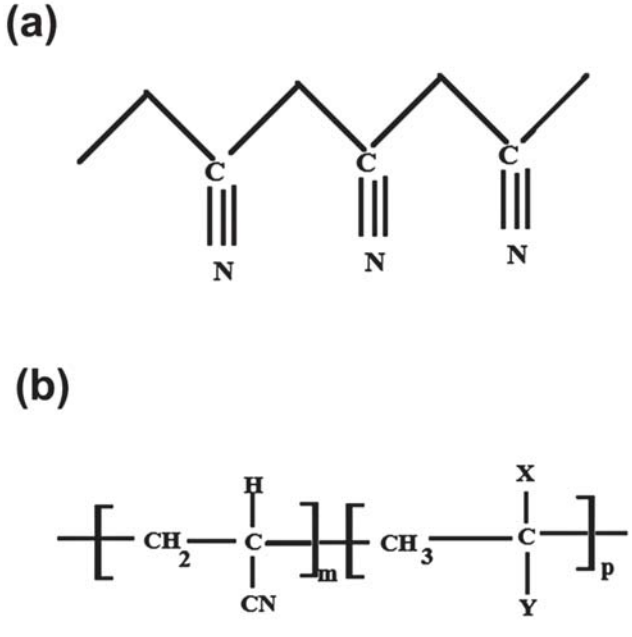

Fig 2: Chemical structure of a. PAN homopolymer, b. PAN copolymer

interactions between nitrile groups, thus increasing the solubility and decreasing the glass transition temperature Tg (~95 ºC) to lower the onset temperature of cyclization during oxidation. PAN copolymer is made by reacting the monomer acrylonitrile (AN) with other comonomers like vinyl acetates, acrylamide, methacrylic acids, alkyl acrylates, itaconic acid[9]. Fig. 2a and Fig. 2b shows the typical structures of PAN homopolymer and copolymer, respectively.

The free radical polymerization technique is generally used to synthesize PAN copolymers. Here, the initiator dissociates first to generate free radicals in presence of heat or activator. The initiator free radical breaks the pi (π) bond in AN and initiates the chain reaction by forming polymer radicals. These radicals propagate the reaction until termination happens, leading to the formation of high molecular weight polymers. The termination of growing polymer chains may happen either by combination, disproportionation, or chain transfer to any suitable agents[10].

Homogenous Solution Polymerization: Otherwise called solution polymerization, in this method, the formed PAN copolymer will be in a dissolved state in a non-reactive solvent such as dimethyl acetamide (DMAc), dimethyl sulfoxide (DMSO), dimethyl formamide (DMF). The initiators are mainly azo compounds. This method is energy efficient since the polymer formed is in the solution phase and can be directly spun into precursor fiber. However, it has certain limitations, such as low conversion and uncertainty in controlling the molecular weight distribution.

Bulk Polymerization: In this technique, AN and comonomers are mixed with initiators without a solvent. The initiators are either azo or peroxy compounds; however, they can also be ionizing radiation or any source of free radicals. The reaction is highly exothermic, so controlling the reaction temperature is a challenge. The bulk polymerisation is not practical in industry, as the reaction is auto catalytic in nature, rapid, highly exothermic and heat removal is very difficult as the degree of conversion is increased.

Aqueous Suspension Polymerization: It is the most viable method for commercial production of PAN copolymer, wherein the precipitated polymer is continuously removed from the reactor after attaining a steady state (see Fig 3). The initiators and activators are mainly inorganic salts such as ammonium persulphate and ammonium bisulphite or sodium persulphate and sodium bisulphite. The method has many advantages, and conversion is fair. A combination of suitable comonomers and polymerization techniques will lead to polymer precursors having predefined molecular weights, narrow polydispersity index, and preferred molecular architectures, facilitating high-performance CFs [11]. PAN copolymers for aerospace-grade CF have typical molecular weights of 95000–300000 $gmol^{-1}$ and molecular weight distribution of 1.5-3.5.

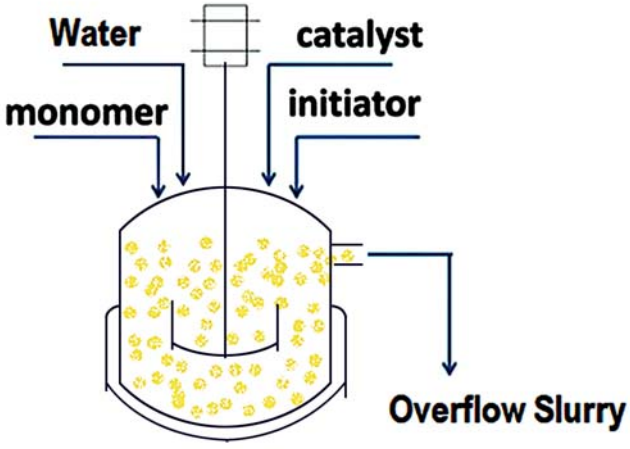

Fig. 3: Schematics of free radical aqueous suspension polymerization

Precursor Fiber

Precursor fiber is made from the polymer using suitable spinning techniques, such as melt or solution spinning, the latter mostly being used. Solution spinning again has different modes like wet spinning, dry spinning, dry jet wet spinning, and gel spinning. Solvents used for different solution spinning techniques are DMF, DMSO, DMAc, aqueous zinc chloride ($ZnCl_2$), aqueous sodium thiocyanate (NaSCN), Nitric acid (HNO_3).

Melt Spinning: The molten polymer mass is extruded through the holes of a spinneret; the filaments are stretched, cooled, and wound on creels. Although melt spinning is cost-effective, it is unsuitable for PAN polymer as it decomposes before melting, and the cyclization reaction of nitrile groups starts even at a lower temperature. Strategies to reduce the melting point of PAN by physical and chemical means, like adding comonomers, water, or glycerol to disrupt the polymer chain's regularity, may favor PAN's transition to a melt state before cyclization[12]. However, melt spinning is promising for lignin-based precursors of CF.

Dry Spinning: The polymer is dissolved in a suitable solvent, and the solvent is evaporated at a temperature above its boiling point in a spin tube with a constant flow of hot inert gas[13]. Further, the fiber is oriented by imparting

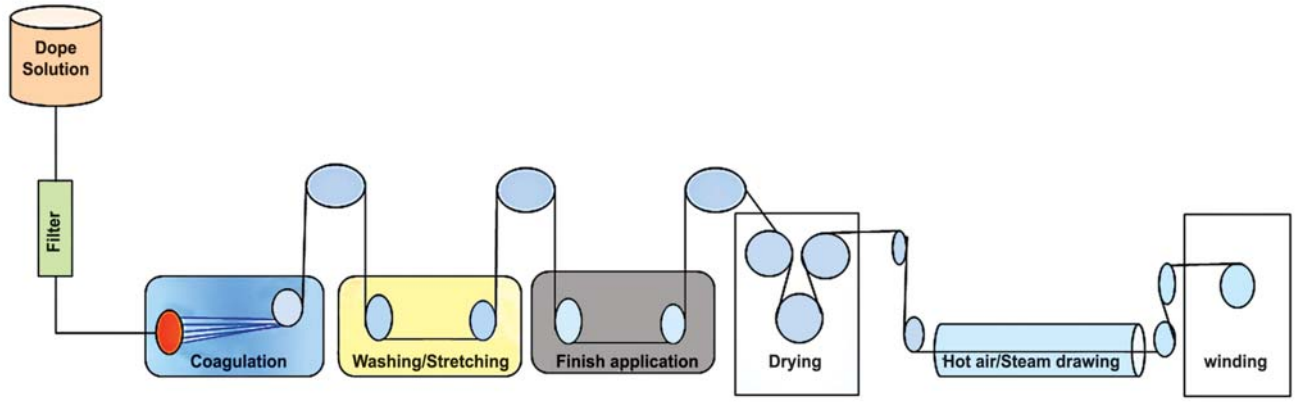

Fig. 4: Schematics of wet spinning process

stretches along the fiber axis. The solvent should be economical and nontoxic, have a low boiling point, and not decompose at the process temperatures. Dry spun precursor fibers usually have a dog bone-shaped cross-section.

Wet Spinning: Wet spinning involves dope (polymer solution) preparation in a suitable solvent, filtration of the dope solution through a filtering media with <1 µm pore size, extrusion of dope through a die or spinneret immersed in a coagulation bath, coagulation of the extruded filaments, washing of the coagulated filament, stretching the filament bundle, drying the fiber tow, hot stretching of the tow either in hot air or steam and finally winding on spools (see Fig. 4)[14]. A coagulation solution is a solvent and non-solvent mixture at a specified weight percent. The coagulation condition controls the filaments porosity, fibrillar structure, and cross-sectional shape. The coagulation conditions must also be carefully controlled to limit the formation of internal pores, surface defects, and skin-core in the fiber. The coagulation solution composition and temperature of the coagulation bath control the coagulation rate of extruded filaments. The spinning filament's cross-sectional shape relates to the volumetric transfer rate of the non-solvent from the coagulation solution into the fiber versus the outward transfer of the solvent from the liquid filament. The fiber skin undergoes coagulation first, thus limiting the fiber's volume. In the coagulation condition, if less solvent diffuses, the fiber becomes non-circular, resulting in a kidney/bean shape.

Dry Jet Wet Spinning: Dry jet wet spinning is a modified wet spinning process. Here, the jet is positioned a few millimeters above the coagulation bath, and the filaments are extruded vertically into the coagulation bath. In this process, the dope temperature and the coagulation temperature are isolated, which allows the use of a higher-than-normal solid content in the polymer solution. Also, the orientation of the filaments is imparted prior to coagulation by stretching the filaments in the air gap. The line speed of dry jet spinning is higher than that of wet spinning. Further fiber processing after coagulation is similar to wet spinning (see Fig. 5)[15].

Gel Spinning: Gel spinning is used to make high-strength and high-modulus fibers. It can form fibers with fewer entanglements compared to conventional solution

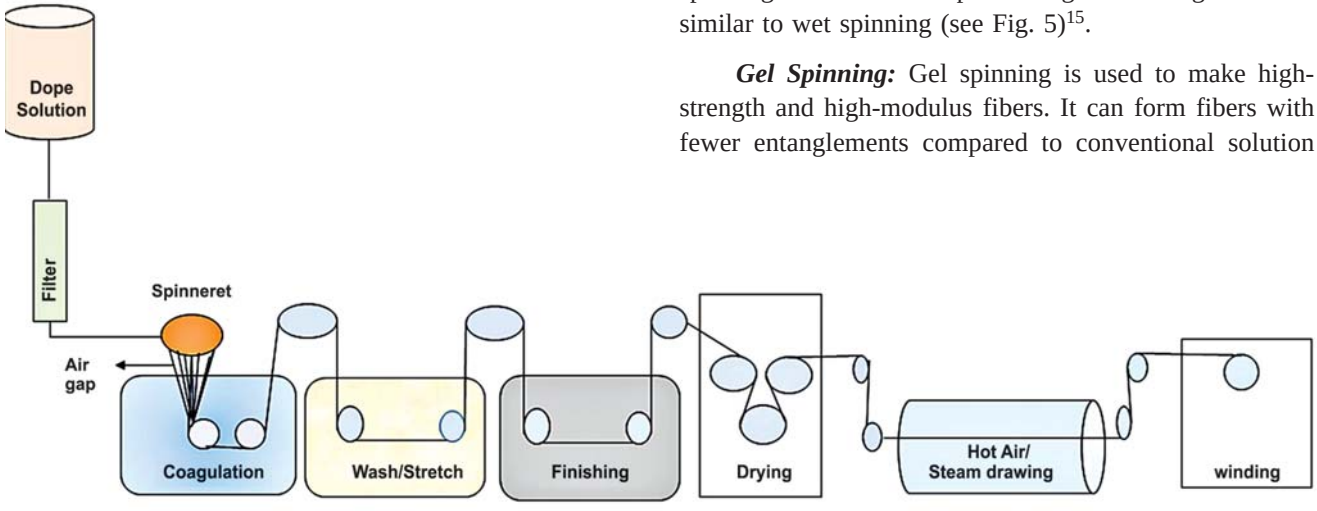

Fig. 5: Schematics of dry jet-wet spinning process

spinning. High molecular weight polymer dope solution can be spun with greater molecular alignment in gel spinning, resulting in high-modulus CF. In gel spinning, the polymer solution is extruded through a spinneret into the air, and then the filaments are gelled by cooling immediately in water or solvent. During gelation, the formation of crystals in the liquid filament and a higher level of entanglements of polymer chains lead to a physically cross-linked "gel" structure. Ultra-high molecular weight polymers are used in gel spinning, and a lower concentration of the dope is used due to the higher level of molecular entanglements[16] increasing solvent use in gel spinning; hence, the processing cost is higher than conventional solution spinning. The gelled filaments are further washed, stretched, and dried before winding them onto the spools.

Most manufacturers use the conventional wet spinning technique to produce PAN precursor fibers. Dry-jet wet spinning is used to spin a dope with higher polymer concentrations and produce CFs with better mechanical properties. The advantages of dry jet wet spinning include the independent spin dope temperature and lesser stretching stress during fiber attenuation compared to the wet spinning process. Additional stretching in the air gap during dry jet wet spinning orients the filament before getting into the coagulation bath. The molecular weight of polymer and polymer concentration in dope for wet or dry jet spinning depends on the desired viscosity of the dope. Melt and dry spinning are not used in commercial PAN precursor production. However, the melt spinning process produces pitch[17] and lignin[18] based precursor fibers. Table 1 summarizes different spinning techniques for the precursor of CFs.

Table 1. Different spinning techniques to make precursor fiber

Technique	Precursor	CF properties	Manufacturers
Melt spinning	Pitch	T.S.: 4.0 GPa T.M: 980 GPa	Nippon Steel Corp, Japan[19]
Melt spinning	Lignin	T.S.: 0.60 GPa T.M: 61 GPa	University of British Columbia, Vancouver, BC, Canada[20]
Wet spinning	PAN	T.S.: 5.8 GPa T.M.: 235 GPa	Mitsubishi Rayon Co. Ltd, Tokyo, Japan[21]
Wet spinning	PAN	T.S.: 6.03 GPa T.M.: 319 GPa	Toho Tenax Co. Ltd., Tokyo, Japan[22]
Dry jet wet spinning	PAN	T.S.: 6.37 GPa T.M.: 283 GPa	Toray Industries, Inc., Japan[23]
Gel spinning-	PAN	T.S.: 5.5-5.8 GPa T.M.:354-375 GPa	Georgia Tech Research Corporation, US[24]

Table 2. Classification of CF based on modulus [25]

Type	T. M. (GPa)	T. S. (GPa)	Tow Size (k)	Application
Standard modulus	220 - 240	3.5 - 5.0	12-50	Automotive, aerospace
Intermediate modulus	280 - 320	5.1 - 7.0	3-24	Pressure vessels, wind energy, aerospace
High modulus	350 - 600	3.8 - 4.5	1-12	Aerospace

Carbon Fiber

CF is a fibrous form of carbon, having various characteristics derived from carbon as a substance and fiber as a form. The characteristics derived from carbon as a substance are excellent thermal resistance, chemical stability, and electrical and thermal conductivity. In contrast, those derived from fiber as a form have excellent strength, modulus, and elongation[25]. Toray introduced the PAN series of CFs into the market in 1971. Since then, many companies have extensively researched and expanded the application of these fibers. Currently, the CF market is undergoing a phase of continuous growth. When PAN series of CFs first came to the market, their application was limited to sporting goods, such as fishing rods, tennis rackets, and golf clubs. However, because of the advanced research and the developmental efforts undertaken by Japanese/U.S. companies, the application area for PAN series of CFs expanded to aircraft, nuclear, construction, transportation, and energy sectors. PAN-based CF accounts for nearly 90% of the world's consumption. The CFs are classified based on their tensile modulus, as listed in Table 2.

Typical heat conversion of PAN fiber to CF involves the following stages (please refer to Fig. 6 for more details): *Thermo-oxidative stabilization*- PAN fibers are subjected to a high temperature between 200°C and 300°C in a flame-resistant oven to produce stabilized PAN fibers.

Pre-carbonization- stabilized fibers are treated in an inert gas (e.g., nitrogen) at 400°C-900ºC to produce pre-carbonized fibers.

Carbonization- Pre-carbonized fibers are again

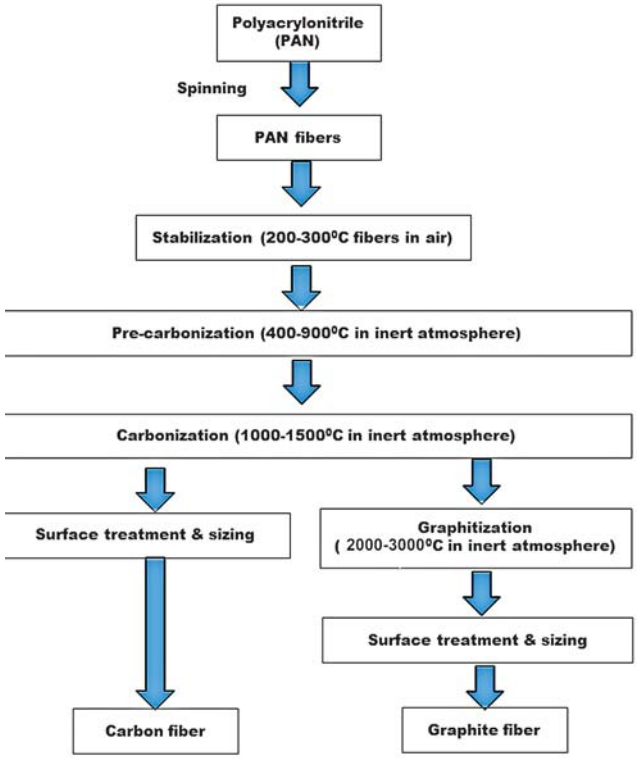

Fig. 6: Heat conversion stages of PAN fiber to CF

treated in an inert gas at 1000°C-2000ºC to produce high-strength, intermediate-grade CFs.

Graphitization- CFs may be further processed in an inert gas (e.g., argon) at a temperature between 2000°C and 3000°C to produce high-modulus grade CFs called graphite fibers.

Surface treatment and sizing- a sizing material (coating agent) is applied to the surface of CFs to produce surface-treated, high-elasticity/high-strength yarns. The surface treatment increases compatibility with epoxy resin during composite making [26].

Resin Formulations for Carbon Fiber Prepregs

The primary functions of a resin are to transfer the load between the reinforcing fibers, keep them intact, and thus protect them from mechanical and environmental damage. Both thermoplastic and thermoset resins are used in the fabrication of polymer composites. Thermoplastic resins include polycarbonate, polyethylene, polystyrene, acrylonitrile butadiene styrene (ABS), polyaniline (PANI), polylactic acid (PLA), poly ether ether ketone (PEEK). The most commonly used thermoset resins are epoxy, phenolic, amino, polyester, vinyl ester, silicone, and polyurethane. High resin content offers good damping properties, i.e., more energy absorption[27]. CF and epoxy resin are the primary choice for making composite structures for aerospace applications. The final properties of cured epoxy resins are affected by the type of epoxy resin, curing agent, and curing conditions[28].

Epoxy Resin Formulation: Cured epoxy resins are hard and brittle and have excellent chemical, thermal, and moisture resistance, low shrinkage, and suitable adhesive and mechanical properties. Because of these superior properties, epoxies are widely used in aerospace, windmills, electronic appliances, automobiles, and marine applications[28]. Fig. 7 shows the typical chemical structure of a cured epoxy resin consisting of diglycidyl ether of bisphenol-A (DGEBA) as a base epoxy resin, methyl tetrahydrophthalic anhydride (MeTHPA) as a curing agent, and (dimethylaminomethyl) phenol (DMP-30) as an accelerator [29]. One or more epoxy resins are mixed with various curing agents, modifiers, and other additives to improve a specific set of targeted properties. Different epoxy resins are available based on the number of epoxy groups, epoxy equivalent weight, and physical and mechanical properties. Bifunctional resins such as bisphenol-A diglycidyl ether (DGEBA), trifunctional resins such as triglycidyl-p-aminophenol (TGPAP), tetra functional resins such as tetraglycidyl-4,4'-diaminodiphenyl-methane (TGDDM), are the most commonly used epoxy resins. These are cured by aromatic amines and suitable for high-temperature applications. Some of the literature references with different epoxy resins, curing agents, and their applications are presented in Table 3.

Curing agents are added to promote the cure kinetics of the epoxy resin by irreversible chemical changes. The molecular structure and glass transition temperature (T_g) of the curing agent significantly influence the cure kinetics of the epoxy resin mixture. Various curing agents such as alkali, amine, catalytic-based, and anhydrides are used depending upon their compatibility, chemical composition, and cure kinetics with the epoxy resin[34]. Curing agents

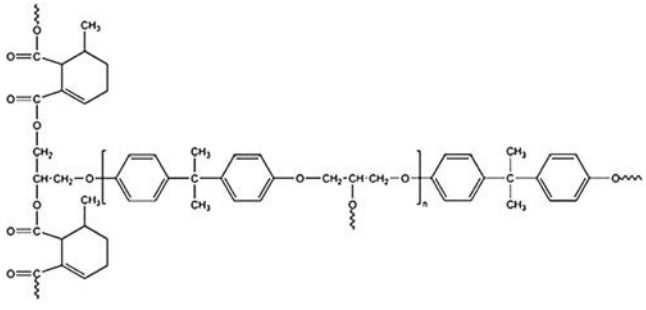

Fig. 7: Typical chemical structure of cured epoxy resin

Table 3. Various Epoxy Formulations and their Applications

Epoxy resin	Curing agent	Application
N,N,N′,N′-Tetraglycidyl-4,4′-diaminodiphenylmethane (TGDDM)	4,4′ -diaminodiphenylsulfone (DDS)	Aerospace[30]
Tetra functional epoxy (TFTE)	4,4′ -diaminodiphenylsulfone (DDS)	Aerospace[30]
Mono-component epoxy resin (RTM-6)	High temperature cure (180°C)	Aerospace[31]
DGEBA + diglycidyl ether of tetrabromobisphenol-A	Polyether amine (PEA)	Adhesives[32]
Epoxidized Cardanol-formaldehyde novolac resins (ECNs)	Self-cured	Surface coatings[33]

such as 3,3′ diaminodiphenyl sulfone (DDS), 4,4′ dicyandiamide (DICY), diethylenetriamine (DETA) are used based on their compatibility with the base epoxy matrix. The high-temperature cured epoxy formulations are used for aerospace applications. Fig. 8 gives a process diagram of making epoxy resin formulation. The recipe of resin formulation depends on many factors, such as the type of base resin, reactivity between the base resin and the curing agent, and cure kinetics. A typical epoxy resin formulation is presented in Table 4 based on reference[35].

Cure Kinetics: The resin is cured under a specific temperature and period depending on its chemical composition and cure kinetics. A polymer network forms by the crosslinking reaction, the degree of which depends on the reactivity of chemicals, temperature, heating rate, and curing time [36]. If the heating rate is incorrect, the molecules may undergo thermal shock, leading to residual stress and poor mechanical properties of the final composite. Therefore, ensuring that the formulated resin mixture does not undergo any thermal shock during the curing, heating, and cooling control is critical. Advanced computational methods such as molecular simulation and three-dimensional finite element method are now available to provide accurate curing conditions for a given resin system[37]. A novel three-stage curing method was proposed in [38] based on a combination of kinetics simulation and extrapolation for predicting the autocatalytic behaviour of the resin system for improved mechanical properties. A typical curing schedule for a trifunctional epoxy formulation with a heating and cooling rate of 2.5°C and 1.5°C, respectively, is shown in Fig. 9[39]. Recent advancements in cure kinetics predict the curing conditions by modelling and simulation with software [40]. The curing of formulated epoxies is usually done in autoclaves or small electrical ovens, depending on the size of the moulds. Out-of-autoclave (OOA) methods such as resin transfer moulding (RTM), vacuum-assisted resin transfer moulding (VARTM), same qualified resin transfer moulding (SQRTM), and balanced pressure fluid moulding are now available for the rapid curing of the epoxy/CF composites [41]. Due to rapid curing, OOA methods provide significant advantages such as reduced cycle time, lower cost, and lesser gas emission.

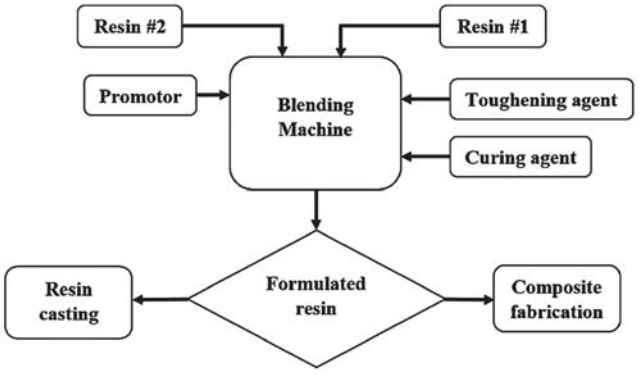

Fig. 8: Process diagram of making epoxy resin formulation

Table 4. Typical epoxy resin formulation

Composition	Chemical name	Basis	Recipe	Batch size
Base epoxy-1	N,N,N′,N′-Tetraglycidyl-4,4′ -diaminodiphenylmethane (TGDDM)	60 wt.%	R1	
Base epoxy-2	Triglycidyl-p-aminophenol (TGPAP)	40 wt.%	R2	R1+R2+R3+R4
Curing agent	(4,4'-diaminodiphenylsulphone, DDS)	50 wt.% w. r. to the base resins (1+2)	R3 = 50 wt.% of (R1+R2)	
Modifier	Polyether sulphone (PES)	10 wt.%	R4 = 10 wt.% of (R1+R2)	

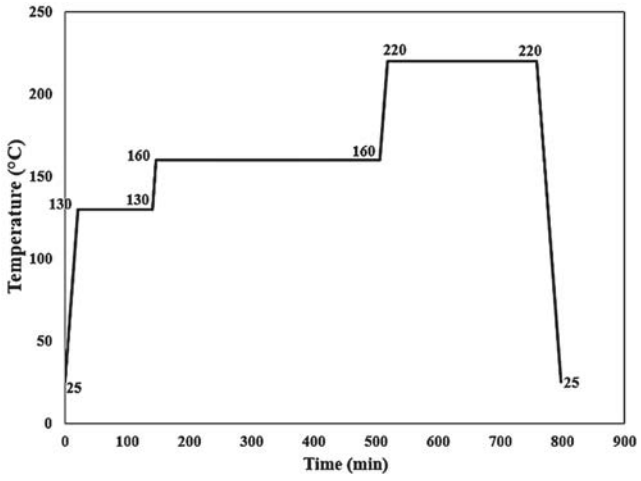

Fig. 9: Typical curing schedule of a trifunctional epoxy resin system

CF Prepregs

Prepregs are laminate composites made of fibers/ fabrics pre-impregnated with partially cured thermoset resins or thermoplastic[42] . Thermoset resins provide higher thermal and chemical resistance and better mechanical properties. The thermoset prepregs market may reach USD 91.4 billion by 2028[43]. Analysts predict a significant increase in the global CF prepregs market, with a projected rise from USD 9.7 billion in 2022 to USD 18.9 billion by 2027[44]. Prepregs result in less wastage during component fabrication. Here, fiber-to-resin ratio and ply thickness are precisely controlled, giving uniformity and repeatability. The curing time is also reduced due to the partial curing of the matrix. Prepregs also reduce the health and safety risks associated with handling resin. Void reduction, low thermal expansion coefficient, and enhanced vibration-damping characteristics are other advantages of prepregs. However, thermoset resins need to be stored at sub-ambient conditions because of their shorter shelf life, and the prepregs are costlier than the corresponding resin-fabric moulded parts. Thermoplastics have the advantages of fast processing, recycling, excellent flame, smoke, and toxicity properties, and unlimited shelf life [45]. These show good chemical and impact resistance, low moisture absorption, and environmental resistance[46]. The global thermoplastic composites market is thriving, reaching USD 22 billion in 2022, and is expected to hit USD 32 billion by 2025 [47]. However, high cost, high melt viscosity, and corresponding high processing temperatures are the disadvantages of thermoplastic prepregs. Ready-to-use thermoplastic sheets are available in different forms, e.g., semipregs, prepregs, and organosheets[48].

Manufacturing of CF Prepregs: Fiber impregnation is essential in producing good quality prepregs. The pressure drop across the fiber bundle thickness controls the impregnation [49]. Advanced machinery ensures uniform distribution of resin throughout the fibers. The consistency and accuracy of impregnation determine the final product's mechanical and thermal properties. The interfacial adhesive force between fiber and matrix should be higher than the cohesive force of the matrix itself to make good prepregs [50]. Wet, dry, and multi-scale fiber surface modifications improve the interfacial properties[51]. In the subsequent section, we discuss different methods of fiber impregnation.

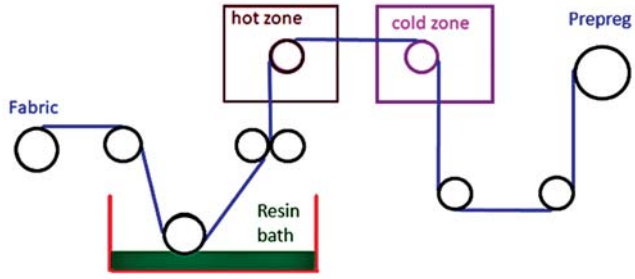

Fig. 10: Schematics of manufacturing CF prepregs via solvent dip method

Hot Melt Method: While the hot melt method requires specialized equipment, it offers a direct coating of molten resin onto fibers, resulting in excellent fiber-matrix adhesion. Importantly, no solvent is used, enhancing its safety profile. Moreover, the production rate is commendably high, making it a reliable and efficient choice for fabric prepreg production.

Film Calendaring: The film calendaring method involves the precise formation of a resin film on paper, either by hot melt or solution. This thin film is then meticulously transferred by passing the fiber yarn between two films and calendaring using heated rollers. The pressure applied by the rollers ensures a partial impregnation of the fiber web, resulting in a prepreg of

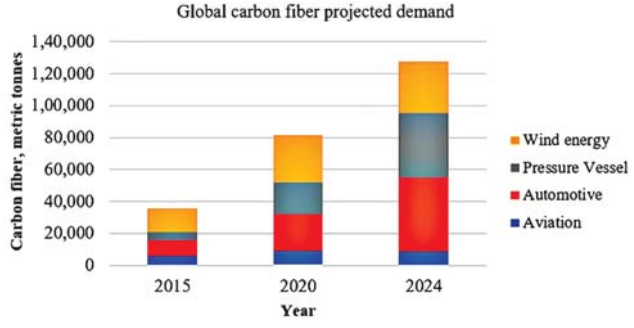

Fig. 11: Global CF projected demand

superior quality that can be wound on a spool or cut into sheets.

Solvent Dip Method: Fabric prepregs are made from fibers pre-impregnated with partially cured resin. It involves dissolving the resin in a solvent of up to 50% solid content and dipping the reinforcement into the solution or spraying the resin solution onto the fabric. Excess solvent is evaporated from the resin, and then the resin is partially cured in the B-stage. The prepreg is then wound onto spools along with release films or cut into sheets (Fig. 10) to enable easier handling. The solvent dip method has the advantage of good fiber-wetting but is associated with health hazards because of the volatile solvent and the longer processing time.

Processing Parameters for Prepregs Manufacturing: Optimum tension on the fibers, ensuring minimal fiber entanglements, is the primary process parameter in prepregs making. The drape value should be within specified limits, while the fiber bundles should be spread uniformly, covering 80-98% of the target width of prepreg and subsequently impregnated with resin[52]. Several factors significantly impact prepreg processing. For solvent-based methods, temperature, pressure within the impregnation bath, production line speed, and fabric tension are crucial. Similarly, hot-melt processing relies heavily on factors like melt temperature, pressure applied by the impregnation rollers, and their rotation speed. Resin tackiness plays a significant role and increases with increasing line speed and fabric tension, while void content decreases with decreasing line speed and fabric tension and increases with increasing impregnation temperature and pressure [53]. Therefore, the process conditions should be set judiciously to optimize the tackiness and minimize the void content.

Resin Content: Resin content determines the final properties of the composites, like strength, stiffness, and weight. It is controlled by adjusting resin viscosity, resin bath or film thickness, line speed, tension to prepregs, and removing excess resin from the fabric. The gap between rollers during impregnation, solvent content, and line speed affect the resin content for solvent-based prepreg processing[54]. Increasing resin viscosity is more effective in increasing resin content than increasing impregnation velocity[55]. Gas transport pathways formed by incomplete prepreg impregnation lead to products with fewer voids during out-of-autoclave processing[56]. The degree of impregnation (DOI) is calculated from the ratio of fibers surrounded by resin and the total volume of fiber regions. Interestingly, reducing the DOI leads to lower void contents [57]. Partially impregnated prepregs lead to void-free panels, whereas full impregnation creates voids exceeding 5%. Research suggests a 60% impregnation level is ideal for void-free results.

Degree of Cure: The pre-curing or conversion of prepregs, is a key determinant of volatile content and prepreg flow. This process is regulated by adjusting the temperature, line speed, and catalyst of the B-stage process[58]. The degree of cure, a critical factor, influences prepregs' tack, drape, and flow properties. Tack is low for both very low and high degrees of curing. At low pre-cure, matrix viscosity and liquid strength are low, while at high pre-cure, resin wettability is less[59]. Other process parameters specific to the resin system and equipment may also impact prepreg manufacturing. Hence, it is imperative to optimize processing parameters based on the desired quality and performance of prepregs. Various analytical techniques such as differential scanning calorimetry, β-ray technology, and non-destructive techniques like IR spectroscopy and micro-computed tomography (Micro-CT) are employed to assess prepreg quality[60].

Process Parameter Optimization in Thermoset Prepreg Laminate Preparation: Process optimization for laminate preparation is crucial to achieve desired composite properties like strength, stiffness, and void content. The most critical process parameters are temperature, pressure, resin content, and cure time. Out-of-autoclave processing relies on an isothermal dwell at 130°C for 30 minutes to ensure sufficient resin infiltration into the dry areas[61]. Increasing the cure temperature accelerates the cure but can lead to thermal degradation. TMDSC evaluates the degree of cure and glass transition and correlates them to curing conditions[62]. TMDSC resolves the total heat flow into reversing and non-reversing components and helps to identify the glass transition temperature. Applying high pressure ensures good fiber-resin interaction but can also cause fiber damage. Hence, pressure optimization is required to minimize voids and improve fiber integrity.

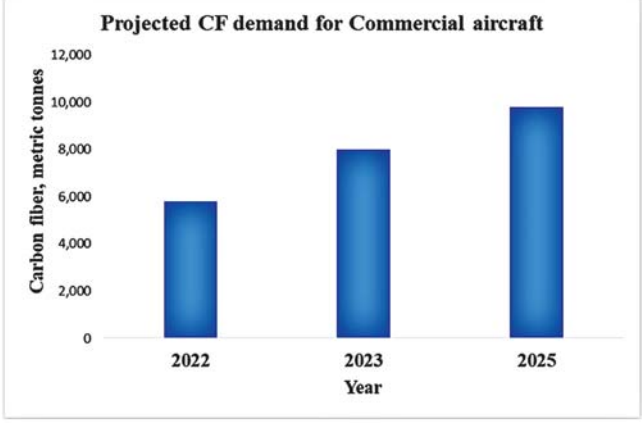

Fig. 12: CF demand forecast for commercial aircraft

Summary and Future Outlook

This review covers different aspects of making precursor material for CFs and manufacturing CF prepregs thereof specific to aerospace applications. As mentioned, CF composites have a higher specific strength than traditional metallic materials, thus lowering aircraft weight and fuel consumption and significantly reducing carbon dioxide (CO_2) emissions. Aircraft manufacturers are inclined to use CFRP composites in their fleets, ensuring a considerable demand for CFs over the next 20 years or until new materials with equivalent or better properties are invented. However, the use of CFRP composites in aerospace structures is still in the early phases because of its high production costs and the need for in-depth knowledge of composite structures, materials, and tooling. Alternative economic precursors with simple processing technologies may lead to the production of low-cost CF.

Composite manufacturers are looking to increase the use of CFRP composites in new areas with innovative, sustainable, and energy-efficient production methods. The Green Alliance, UK, has warned that CF is one of the several novel materials that could create waste problems in the future, and swift action should be taken to recycle and reuse the CFs [63]. Over the past 35 years, over 16000 passenger and cargo planes have been retired worldwide[64]. The usual disposal routes for end-of-life composite structures are landfills or incineration, a poor waste management strategy that pollutes the environment. Therefore, the second life for CF composites, reusing and recycling, should be managed sustainably and profitably to reduce the manufacturing requirement of new precursor materials (energy-intensive process), thus reducing costs and empowering an eco-friendly strategy. Using recycled CFs in a secondary market other than the aerospace industry may lead to sustainable and profitable recycling planning. The replacement of thermosets with thermoplastics is a promising route for the recycling and reuse of composite. A significant effort has been devoted to developing high-performance thermoplastic composites for highly demanding aerospace applications. Also, with the expanding application of CFRP composites from aerospace to non-aerospace industries such as automotive, wind energy, rail, and ship buildings, i.e., a large volume of usage for economical and sustainable applications has a broad future for CF composites.

Issues and Perceptions of Current CF Manufacturing: The United States, Japan, and European economies are the current hubs of CF manufacturers and drivers of demand [65]. However, it is crucial to note that new competitors such as China, Korea, and Russia are emerging with a focus on capturing a larger share of the CF markets. This shift in the industry landscape demands our attention and strategic planning. As always, the aerospace and defense sectors remain the driving forces for the development of CF industries. Different PAN series of CFs are developed based on process refinement and production expertise. However, the resultant properties of CFs are far from theoretical limits. CFs produced are still less than 10% of the theoretical strength of the carbon-carbon bond, and the modulus is about 30% of the theoretical modulus for PAN-based CFs. The defect control capability at the nano level, improving the fracture toughness, and optimizing the interface properties are the major challenges to making high-performance CFs. Optimizing the microstructure can improve CF strength by decreasing its flaw size and density. So, controlling precursor morphology and tuning processing conditions for each precursor system should be explored continuously[66].

PAN-based CFs, as we know, satisfy the demands of most aerospace applications. This is a testament to the crucial role of the aerospace and defense sectors in driving the development of CF industries. However, the primary concern is the high capital-intensive plant construction, which possesses excellent economies of scale and demands stringent quality control capabilities for product certification in aerospace applications. The strict performance requirements of CF for aerospace structures lead to tighter manufacturing tolerances and higher costs than those associated with other CF applications. The product certification for military aircraft may take several years, and for commercial aircraft, more than a year.

Recent Developments and Trends in CF Technology: Commercial aviation segments currently dominate CFs aerospace market, with large passenger and cargo jets producing nearly 77% and military combat and transport aircraft making up the remaining 23% of the total CF usage in the aviation sector. However, the future is promising, as Fig. 11 illustrates the overall projected CF demand in various sectors. The demand for CF in the aviation sector is set to grow at a CAGR of 16% [67]. Fig. 12 provides a glimpse into the CF demand forecast for the commercial aircraft segment. CF composites per Airbus aircraft (A380) require nearly 35 tons for its primary/secondary structures, reducing weight up to 20% compared to conventional aircraft [68].

In order to expand the CF market, it is crucial to focus on a cost-effective production process, improvement of matrix resin performance, and innovation of composite production technology, along with the enhancement of CF

performance. Recent trends in aerospace-grade CF technology are encouraging, as they are geared towards reducing the cost of CFs by various approaches, such as innovating fiber machinery for large tow processing and improving the speed of the stabilization process. At present, tow sizes suitable for aerospace-related applications have been reliably scaled up to 24k using advanced processing machinery. In today's scenario, advanced analytic techniques and computer modeling also significantly assist fiber research [69]. The area of CF is still providing many new opportunities for research and development-focused and innovation-driven firms and entrepreneurs, inspiring a wave of new ideas and solutions.

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