



Polymer composite for antistatic application in aerospace

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ABSTRACT

The phenomenon of static electricity is unpredictable, particularly when an aircraft flying at high altitude that causes the accumulation of static charges beyond a threshold value leading to the failure of its parts and systems including severe explosion and radio communication failure. The accumulation of static charges on aircraft is generated by the virtue of interaction between the outer surface of aircraft and the external environmental attributes encompasses air particles, ice, hail, dust, volcanic ash in addition to its triboelectric charging. In the recent years, advanced polymer-based composites or nanocomposites are preferred structural constituents for aircrafts due to their light weight and comparable mechanical properties, but such composite systems do not render low impedance path for charge flow and are subsequently vulnerable to effect of lightning strike and precipitation static. In this context, it is essential to develop conductive composite systems from non-conductive polymer matrix by nanofiller embodiments. The advent of carbon-based nanocomposite/nanomaterials have adequately addressed such issues related to the nonconductive polymer matrix and further turned into an avant-garde genre of materials. The current review envisioned to illustrate the detailed exploitation of various polymer nanocomposites in addition to especially mentioned epoxy composites based on carbon fillers like carbon black, carbon nanotube (single walled carbon nanotube and multi walled carbon nanotube) and graphene the development of antistatic application in aircraft in addition to the static charge phenomenon and condition for its prevalence in avionic systems.

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1. Introduction

The accumulation or development of “static electricity” over a dielectric surface is not a modern phenomenon in fact, it was first observed by Greek scientist Thales of Miletus (624 BC to 547 BC) who stated that the amber attracts small dust when rubbed with animal fur [1]. Later, London based William Gilbert (1544–1603) discovered that other materials could be also charged to attract different object apart from amber and he coined attractive force as “vis electrica” while repulsive force as “noelectriks”. Pionteck and Wypych elucidated that though, he did not elaborate the real attributes of electrostatic attraction but successfully distinguished it from magnetic attraction [1]. In 1757, the development of triboelectric series by J.C. Wilcke marked a significant advancement in

the field of electrostatics [2]. It was noted that when two materials were rubbed together, they produced opposite charges and once which preceded in series turns positively charged. Since then, number of triboelectric series has been constructed but wide inconsistency was observed in all series because electrostatic phenomenon is a surface attribute and largely depends on surface properties of materials, influenced by its environment, preparation condition and previous history [3]. Luttgens and Wilson [4] explained static charge phenomenon based on Helmholtz [5] hypothesis of double layer charge which is elaborated as follows: when two materials have firm contact with respect to each other and subsequently separated, a double layer charge was produced at the interface, called as triboelectrification. In this view, the surface of material with lower work of function developed the positive

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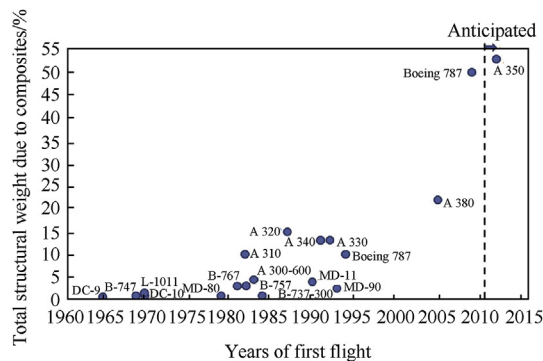


Fig. 1. Chronological utilization of composite in aerospace application [6].

charge while the other surface with higher work functions established with negative charge.

The electrostatic charges developed on any surface is not stable and attractive or repulsive force disappeared with time depending on material type, environmental stimulus and other factors. Based on such electrical activity, materials are mainly divided as insulator (conductivity $\sigma = 10^{-22}$ to 10^{-12} S/cm), semiconductor ($\sigma = 10^{-9}$ to 10^{-2} S/cm), conductors (or metals $\sigma = 10^2$ S/cm) and superconductors ($\sigma = 10^{20}$ S/cm) [1]. As per the ASTM standard ASTM D1711 – 15, volume resistivity, surface resistivity and relative permittivity are the important terminology to define the insulating property of any materials. Based on definition, most of the polymers are categorised in terms of insulators with the conductivity range of 10^{-12} to 10^{-20} S/cm, where all the electrons are localized in covalent bonds without any movement. Since the utilization of polymer-based composites systems in aerospace application is augmenting at enormous pace, therefore, conductivity is gleaned as an essential parameter in the context of static charge phenomenon. This review article elaborates the exploitation of various polymer composites and their fillers to transform the insulating attributes of polymer into conductive nature. Current review also exploits deliberation on various pathways which contributes to electrostatic charge phenomenon on aircraft and their plausible mitigation methodology by incorporating conductive fillers in intended polymer composites.

2. Composite for aerospace application and static charge phenomenon in polymers

In the recent years, the use of composite in the aviation sector for primary structures is rapidly increasing which is exponentially augmented further from last decades as delineated in Fig. 1. For instance, the Boeing 787 Dreamliner have exploited the utilization of approximately 50% of composite materials which has been launched in 2007 compared to the 11% of composite materials in Boeing 777 launched in 2000 [6]. The substantial increase in the utilization of composite materials based on thermoplastic and thermoset matrix are expected to impart light weight, high static strength, sufficient fatigue performance, high fracture toughness and damage tolerance, high impact energy, multifunctionality, easy design and fabrication techniques [6–8]. Rana and Fanguero have classified advanced composite in various application based on requirements, structure and fabrication technique as elucidated in Table 1 [6].

It has been widely reflected that the available composites or nanocomposites can render most of the above explained functionality, however, the selection of matrix and reinforcements become major process constraints to develop a composite for desired applications [17]. Mangalgiri has provided the detailed deliberation on various composite matrices exploited for aerospace application and elaborated that epoxy accounts for maximum compared to total composites utilized in the aerospace application [18]. He has reported that the research work in the recent years has extensively focused on thermoplastics as a primary matrix for various applications related to the thermal, electrical and mechanical functions while thermosets have been progressively explored for high end applications where there is temperature, strength, wear and damping barrier limitations experienced by the thermoplastics. The detailed comparison on various matrix system has been provided in Table 2. It is essential to mention here that the details provided here on the composite for aerospace application is only brief contemplation which can be further elaborated in above stated references and elsewhere [19,20].

Polymers matrices in aerospace application are in essence insulators which is further turned into conductive by incorporating conductive fillers depending on their application and requirement.

Table 1
Advanced composites and their properties [6].

Advanced Composites	Properties	Materials
Laminated composites	Enhanced fracture toughness, damage tolerance, improved impact properties	Carbon fiber, para aramid fiber, E-glass and other textile-based system in various architecture
Sandwich composite	Light weight; High bending stiffness; Cost effective; Thermal insulation; Noise insulation; Vibration damping [9]	Matrix- Epoxy, unsaturated polyester and vinyl ester, polyetherimide (PEI), polyetheretherketone (PEEK), polycarbonate, E-glass, S-glass, HM-carbon, HS carbon, Boron, Aramid
Braided composite	High shear torsional strength and stiffness; High transverse strength and modulus; Damage tolerance and fatigue life; Notch insensitivity; High fracture toughness; Possibility to develop complex and near-net-shape composites [10]	Epoxy, polyester, polyetheretherketone (PEEK), polypropylene, Nylon 6,6, Carbon T300 [11], Carbon T600 [12], Aramid
Auxetic Composite	High shear modulus; Synclastic curvature; High damping resistance; High fracture toughness; Enhanced crack growth resistance; High energy absorption capability [13]	Polyurethane, polytetrafluoroethylene, ultra-high-molecular-weight polyethylene, epoxy, carbon fiber, glass fiber
Ceramic Matrix Composite	High fracture toughness; Resistance to catastrophic failure; High strength; Light weight; Low thermal expansion; Ability to withstand high temperature, High oxidation resistance [14]	Aluminosilicate 1720, Glass ceramics nonmachinable 9606, SiC HP, Mullite, Si ₃ N ₄ HP, boron fibers, glass fibers, oxide fibers
Metal-Matrix Composite	High transverse strength and stiffness; High ductility and fracture toughness; High temperature resistance; High fire resistance; High electrical and thermal conductivities; No moisture absorption; Better radiation protection [15]	Aluminium, magnesium and titanium, glass fiber, boron fibers
Nanocomposite	High surface area and better interface, smaller defects, Low volume fraction required for property enhancement [16]	Epoxy, phenolic, polyimide, bismaleimide, Polyether ketone (PEK), Polyether ketone ketone (PEKK), elastomers, single and multi-walled carbon nanotube, graphene etc.

Table 2
Advantages and disadvantage of various polymer matrix utilized in aerospace applications ^a.

	Thermoplastic	Thermoset	Elastomers
Advantages	Nonreacting; no cure required Rapid processing High ductility High fracture toughness High impact resistance Absorbs little moisture Can be recycled	Low processing temperature Low viscosity Good compression properties Good fatigue resistance Good creep resistance Highly resistant to solvents Good fibre wetting for composites	Low processing temperature High ductility and flexibility High fracture toughness High impact resistance
Disadvantages	Very high viscosity High processing temperature (300°C–400 °C) High processing pressures Poor creep resistance	Long processing time Low ductility Low fracture toughness Low impact resistance Absorb moisture Limited shelf life Cannot be recycled	Long processing times Poor creep resistance Low Young's modulus Low tensile strength

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The phenomenon of static charge electrification in polymers can be expounded based on classical theory of average material's properties [4] and Baytekin et al. "mosaic" hypothesis [22]. The description of classical theory evolves in the context of work of function which is defined as energy required to remove an electron from any surface to infinity. As stated earlier, when two materials are in contact (physically or frictional) the materials with low work of function acquire the status of donor (positively charged) and other material turned into acceptor (negatively charged). The state of equilibrium is achieved when the potential difference between the work of function equals to the potential difference of formed double layers of opposite polarity. In such state, capacitance of formed layer becomes very high which gleaned potential difference at the interface in order of millivolts. Subsequently, the separation of contact surface render dramatic reduction in capacitance which further lead to the high potential difference between contacted surfaces. Polymers exhibit nearly insulating characteristics without any conducting reinforcement and therefore, electron transfer mechanism for contact charging is not always applicable [23]. In essence, such contact charge transfer can be effectively explained by electronic basis which predominantly depends on surface state of insulator. The simplified model based on surface state of insulator by assuming uniform surface density per unit area [24] which unambiguously elucidated that the insulator surface is proportional to the work function of metal, resulting from effective capacity of surface instead of critical separation between metals and insulator. In the context of insulator and insulator contact, the analogous principle can be effectively extended to contemplate the contact charging phenomenon. In addition, contact charge phenomenon can be also explained by ion transfer, especially for the polymer which contains ionic structure or ionic ingredients. It is evident that when antistatic agents bloom out on surface, the charge transfer is predominantly explained via ionic charge transfer mechanism which is dominated by number of factors including the degree of ionic dissociation, the mobility of ions and the relative stability of positive and negative ions [23]. The required for ionic dissociation of two ions is inversely proportional to the static dielectric constant of the polymer in addition to the temperature and the Boltzmann factor [25]. Seanor has demonstrated that the ionic dissociation can be accelerated in the availability of water and subsequently promotes its mobility at higher level [26]. Water is also expected to act like an electron donor and charge carrier [26]. In contrast to the classical hypothesis of static charge, Baytekin et al. demonstrated that the static electrification cannot be explained by spatially homogeneous surface attributes [27] and uniformly charged contacted materials [28] but requires chemical and possibly

micromechanical approach at and near the contacted polymer surface. They have contemplated that the surface of non-elemental insulators are random mosaic of positively charged and negatively charged nonoscopic dimensions which are universal in nature and possesses ability to accommodate significantly more charge per unit area [22]. It is concluded that the polymeric materials exploited in aerospace application requires lower surface resistivity to effectively transfer the accumulated charges without causing danger to the external environment.

3. Static electrification in aerospace composites

The term static electrification is defined by L.B. Loeb which includes all processes that produces segregation of positive and negative electrical charges by mechanical action, operated by contact or impact between solid surface or in the rupture or separation of solid or liquid surface by gases or otherwise introducing ionized gases. Such electrification phenomenon is generated by the virtue of friction, contact or tribo-electrification, spray electrification, and electrification in dust, snow or in thunderstorm [29]. He has delineated static electrification in seven broad categories as follows [29]:

- 1) Electrolytic phenomenon: It involves the range of processes from galvanic cell to the formation of Helmholtz double layer at the surface of metal or other substrates when it contacts with liquid usually with high dielectric constant liquid due to electrolytic ion transfer.
- 2) Contact or Volta electrification: This phenomenon is generated between the clean dry metal to metal or metal to semiconductor surface by the virtue of electron transfer between boundaries with different energy level.
- 3) Spray electrification: This phenomenon persists due to the disruption of surface film of dielectric liquid and solution due to the mechanical forces like atomizing and bubbling of liquid and presumably shattering of high velocity got by solid surfaces. It is contemplated as a consequence of intrinsic electrical double layer at dielectric liquid interface and sometime inseparable from Helmholtz double layer in some aspect.
- 4) Frictional or tribo-electrification: This phenomenon is pertinent to the contact and subsequent separation between two dissimilar surfaces including two insulating solids, an insulator and metals or two solids containing same ion in different concentration. It is believed that true frictional electrification occurs when carrier density and energy differ on asymmetrical heating, resulted due to the rubbing.

- 5) Homogeneous or symmetrical charge separation: A small solid particles or liquid droplet separation from similar particles or larger portion or same matter lead to the formation of equal opposite charges with no net charge accumulation of one sign.
- 6) The generation of ions and electrons by the electrical occurrence in gases in contact with solids or in flames where these ions and electrons are segregated and separated by the mechanical forces resulted from the mass motion of gases or solids.
- 7) In some circumstances, crystals of dielectric surface are polarized by electrical or mechanical forces and strong frictional interaction abrades the section of polarized surface. Thus, piezoelectrification or pyroelectrification of quartz can lead to charging by the virtue of charge removal from the surface due to the heat induced strain. Such phenomenon is mainly pertinent to the aqueous layer and liquids.

Referenced from above classification of static charge accumulation phenomenon, the increasing number of polymer-based composites in aircrafts lead to the development of huge static charges and consequently, a potential hazard to navigation electronics and radio communication. Gilgliotti had reported that the accumulation of static charges and subsequent discharge can produce following consequence on aircraft [30] which also illustrated in Fig. 2:

- 1) Corona effect: formation of bright edge near the extremities of plane (tips and tails). In general terms, Giacometti and Oliveira Jr had elucidated that corona is self-sustainable, non-disruptive electrical discharge which develops when sufficiently high potential difference exists between asymmetric electrode such as fine wire or a point and a plate or a cylinder [31]. They have reported that the transformation into ionized air from insulating characteristic of air is related to the high electric field near emitting electrode which further results in driving of ions towards low-field electrode. The threshold of such corona discharge mainly depends on availability of free electrons, exists in different regimes. The autostabilization regimes are contemplated to the condition where positive and negative corona is pulsating just above the threshold. When the voltage augments the positive corona exhibits glow discharge while negative charge reaches to the regular Trichel pulse's regime that is also turned into continuous glow discharge at extremely high voltage.
- 2) Streamering: formation of electric charge along the insulating materials like canopy which discharge to the extremities
- 3) Generation of spark between the parts which are mechanically connected but electrically isolated

Nanevicz elucidated that static charge accumulation on aircraft may evolve due to the precipitation static, engine exhaust charging,

and exogenous charging [32]. The precipitation charging occurs when aircraft aviate through dry precipitation of clouds in the form sleet, hail or snow. The aircraft acquires the preponderance of potential gradient at its surface compared to external environmental condition when impacted with such precipitation in the cloud, coined as precipitation static or P-static [33]. The P-static is a continuous phenomenon which last for several second to minutes, but it is rarely observed during rain. While the engine exhaust charging is speculated at the lower attitude and hypothesized that the exhaust may contain positively charged ionized particles which impart negative charges on aircraft [32]. The exogenous charging evolves when aircraft aviate through a positively charged region of clouds [32]. The electrification of aircraft can also be experienced due to the lightning under cumulonimbus clouds and potential hazards are essentially related to the discharge of such accumulated static charge, called as electrostatic discharge phenomenon (ESD). ESD is contemplated as an event, in which finite amount of charge is transferred from one object to another in a very short period of time [34]. Sweer et al. have reported various possible zones in Boeing aircraft that can experience different level of lightning strikes as elucidated in Fig. 3 [35]. Zone 1 indicates an area likely to be affected by the initial attachment of a strike. Zone 2 indicates a swept, or moving, attachment. Zone 3 indicates areas that may experience conducted currents without the actual attachment of a lightning strike (Reproduced from open source provided on https://www.boeing.com/commercial/aeromagazine/articles/2012_q4/4/). It is reported that lightning discharges via atmosphere from one cloud to another or directly to the earth surface. Extremely high-power dissipation has been recorded (100 TW) under lightning condition which possesses ability to break the dielectric constant of air (depends on the moisture content) [36]. The prevalence of such extreme condition causes the generation of sufficient electromagnetic field strength to be injected into the electronic equipment of aircraft.

It is evident that ESD induced current possesses ability to render electrical interference to the sensitive circuitry and causes severe explosion and radio communication EMI interference or jamming of Radio-Navigation aids momentarily originating flight safety hazards in aircrafts [37]. Apart from aerospace industry, it is estimated that 35% of microchip damages related incidents are emanated due to improper dissipation of ESD generated energy [34,38]. Conventionally, aircraft structure contained aluminium skin which provided highly conductive path for flow of generated lightning strike. However, in the composite system having dielectric surfaces are preponderance to the accumulation of static charges and subsequently cause severe damages to the aircraft structure directly or indirectly [39]. The utilization of composite structures mainly develops varying interface leading to the escalation possibility of material mismatch between finishes, fasteners and adhesives that causes a dramatic change in electromagnetic capability of exploited materials [40].

The advent of nano technology in the field of polymer composite has demonstrated the ability to circumvent the prevalence of such static charge accumulation and simultaneously provides synergistic feature of polymer and nanocomposite that forge the design of avant-garde genre of materials [41]. Though polymers are inherently insulating in the nature but the nanofiller embodiments like carbon black, carbon fiber, 1-n tetradecyl-3-methylimidazolium bromide, metal particles and nanofiber like polyaniline [41–45] have demonstrated the significant augmentation in conductivity to exceeds the insulating threshold. Though, the detailed evaluation of composite or nanocomposite materials for aerospace application has been extensively exploited [6,17] but utilization of such materials for circumventing static charge phenomenon in structural aerospace application has been least explored in the form of review.

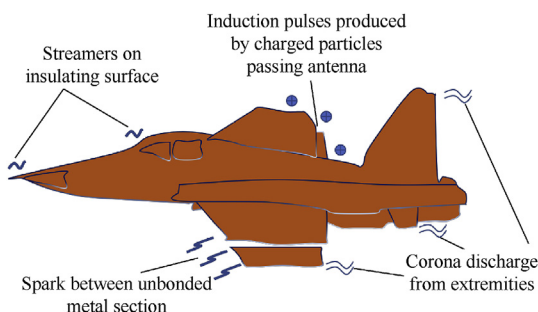


Fig. 2. Effect of static charges on various sections of aircraft.

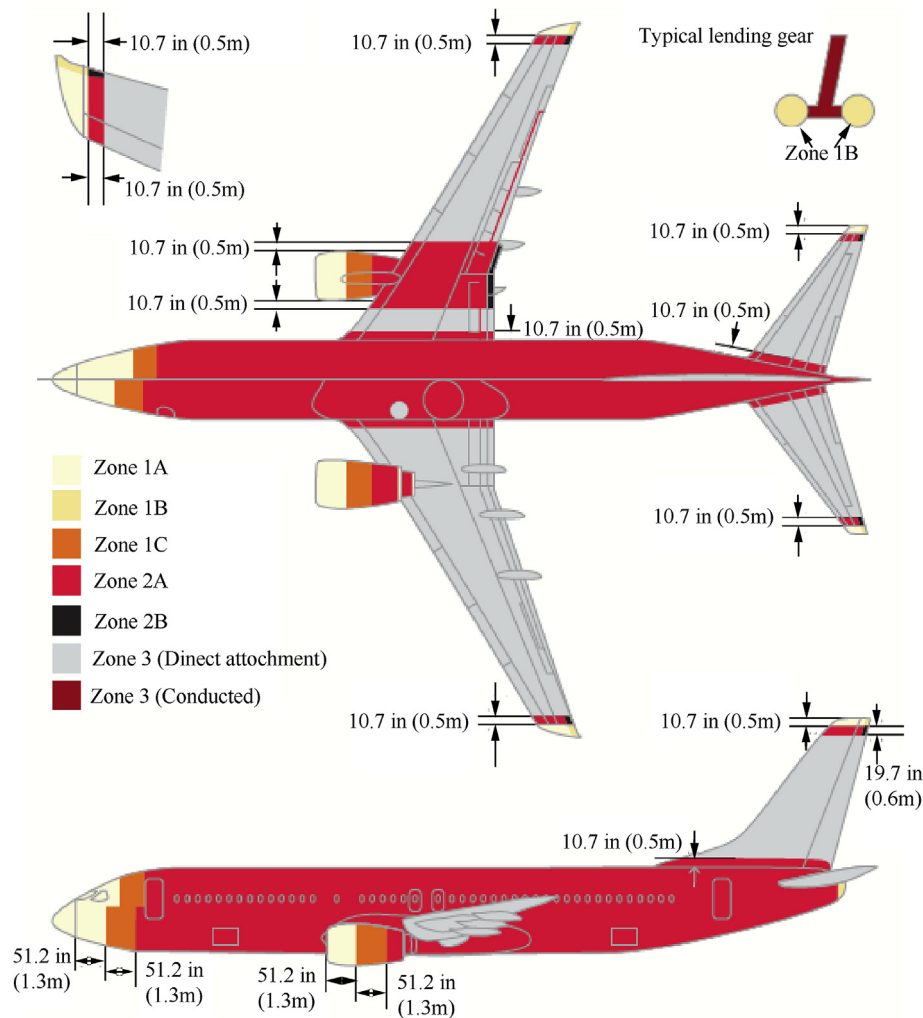


Fig. 3. Airplane lightning zones. Areas of an airplane that are prone to lightning strikes are indicated by zone.

This review article focuses on the recent development of polymer nanocomposite in the field of static discharge phenomenon and its application in mitigating the hazards related to its accumulation. The current abstraction also includes the development of carbon-based nanocomposite for circumventing the electric static hazards and their mechanism of mitigation. Though, extensive literature is available on various aspects of static discharge phenomenon and methodology to evade its effect on composite materials but the detailed contemplation on carbon based composite materials specifically for aerospace application is reported in list. The review article also provides the relevance of percolation threshold phenomenon in developing electrostatic discharge composite. Though, it is too ambitious to evaluate all the published work in the field of static charge application, but we envisioned that the current review renders critical contemplation on currently available composite technology for evading the related electric hazards in aerospace application.

4. Nanocomposite for static charge extenuation

Eric Drexler in 1986 neologized the term nanotechnology as “technology at molecular level that can let us set every atom where we want it to be. We called this ability ‘Nanotechnology’, because it works on nanometric scale, one billionth of meter” [46]. It is widely contemplated that when size of materials reduces below 100 nm,

dramatic changes in properties are perceived which offers significant property enhancement even at low dispersant loading [47,48]. The utilization of such dimensions are profoundly observed in the production of physical, chemical and biological systems as well as integration in larger systems like polymers [49]. In aerospace application, the nanofiller reinforcement is essentially exploited to augment the load bearing capability and synchronously provide functional or the non-structural property to polymer matrix (thermosets or thermoplastics) [50,51]. The reinforced nanoparticles are mainly classified in the three main categories including nanoparticles (metal nanoparticles, ceramics particles), nanotubes (nano fibers, whiskers, nanorods) and nanolayers (clay, layered double hydroxides) [52]. In this context, Baur et al. have demonstrated the utilization of various nanoparticles and its exploration for various avionic applications as illustrated in Table 3.

In the context of static charge extenuation in avionic composite, the embodied fillers in the polymer are sometimes referred as anti-static agents and mainly classified based on their application mode or composition and their chemical structure [53]. Pionteck and Wypych have exemplified that antistatic agents on their application mode can be alienated into internal and external antistatic agents where externally applied moieties are surface active ionic or nonionic whereas internally active agents are exploited for developing conductive pathways in polymeric materials by reinforcement [1]. Based on the chemical structure, antistatic agents can be

Table 3
Inclusion of various nanoparticles for aircraft structure and components ^a.

Application	Nanoparticle inclusions
Cryogenic tanks and optical benches, antenna trusses	High aspect ratio silicate or graphite
Thermal protection system and space structure	Incorporation of high temperature, oxidation resistance fillers like silicates, CNTs and POSS
Charge discharge adhesive coatings	High aspect ratio conductive particle like CNTs, graphite flakes and metals etc.
Electronic and bus compartment enclosure	Highly percolated network of conductive fillers like nickel nanostrands veil and SWNT buckypaper etc.
Composite aircraft extremes	CNT, percolated coatings, appliques, resin or veils
Thermally conductive adhesives, radiators, doublers, electronics boards	Incorporate highly thermally conductive particles (CNTs, metals, etc.) Thermally conductive into resin and optimize structures for heat transfer along adhesives, gaskets, continuous path to heat sink
Aircraft brakes, re-entry components	Thermally conductive and insulating nanofillers within resin to direct heat away from vehicles, missiles protected systems or enhance mechanical properties at high temperature
Membrane structure, damage tolerant structure	CNTs, layered silicate or silica
Propulsion tank and fittings	Functionalized CNT
High temperature composite vehicle, vehicle health monitoring.	Grow high strength nanoparticles such as CNTs from fiber to tailor the interfacial properties as a smart sizing.
Tubular structure	Incorporation nano filled resins with increased toughness at mid-ply via coatings or prepregs.

^a Reproduced with the permission of Cambridge University Press from Baur et al. Challenges and Opportunities in Multifunctional Nanocomposite Structures for Aerospace Applications, MRS bulletin [51].

classified into anionic, cationic, atmospheric (amine oxides and betaine type antistatic) and non-ionic (long-chain amines, amides, sulfur, or phosphorous as heteroatoms). Apart from this antistatic agents are also categorised as conductive polymer (polypyrrole, polythiophene, polyphenylene), poly (phenylene sulfide) or polyaniline and inorganic based agents (alts, metals, semiconductors, carbon black, carbon nanotubes, and other intrinsic conductive or conductive modified particles) [1]. The purpose of antistatic agents is to impart the effective conduction pathways to dissipate the accumulated static charges in polymer-based aerospace composites. In addition to reinforcement driven conductivity, polymer can achieve conductivity through ions originated from small-molecular impurities and electronic conductivity, evolve due to the motion of electrons along the polymer chains. The conductivity of the reinforced polymer nanocomposites predominantly depend on percolation phenomenon, contemplated as random process and distinguishable from diffusion as the particle is considered to be trapped on lattice in percolative walk [54]. Microscopically, percolation is defined as a point at which, first continuous macroscopic length appears in polymeric system and beyond this critical volume fraction of filler the overall resistance of the polymer abruptly dropped to further approach the conductive value of the filler [55]. Percolation threshold and bulk conductivity of the polymer also depends on geometrical parameters of the fillers such as filler size, shape and aspect ratio, dispersion state of filler and method to disperse the fillers in the polymers [56]. The process of charge transfer in filler reinforced composite is advocated to be governed by two different pathways, 1) the injection of charge carriers in polymer due the conductive reinforcement, 2) the movement of charge carriers through hopping, tunnelling, ballistic transport, ballistic transport, diffusion or metallic conduction [57]. Q. Xue and J Sun have reported that the simplified equation proposed by C-W Nan as follows [56,58]:

- 1) The conductivity of the nanocomposite (σ) is determined by the conductivity of the filler (σ_1) by utilizing Eq. (1) when the volume fraction of filler (f) just exceeds to the percolation thresholds (f_c). In this condition, the reinforced filler develops continuous percolating path in polymer matrix.

$$\sigma \approx \sigma_1 (f - f_c)^\alpha \quad (1)$$

Where α is the critical component, depends on the connectivity of the system.

- 2) When volume fraction of filler is less than percolation threshold, the matrix restricts the development of percolative pathways and therefore the conductivity of the composite is given by Eq. (2) as follows:

$$\sigma \approx \sigma_2 (f - f_c)^{-\beta} \quad (2)$$

Where σ_2 is the conductivity of the matrix and β is the critical component, depend on dimensionality of the system.

- 3) When the relationship between the volume fraction of filler and percolation threshold is given by $|f - f_c| \rightarrow 0$, the conductivity of the composite is provided by Eq. (3)

$$\alpha \cdot \alpha \cdot (\alpha_2)^\gamma (\alpha_1)^{1-\gamma} \quad (3)$$

Where $\gamma = \alpha/(\alpha + \beta)$.

Carbon based antistatic agents such as graphene, carbon nano tube (multi walled carbon nanotube and single walled carbon nano tube), carbon nanofiber and graphene oxide are extensively employed for the extenuation of static charge accumulation on polymeric surface. Carbon black (CB) is a traditional nanofiller in epoxy resin as electrostatic or antistatic agent in aerospace application which is produced by the incomplete combustion of aromatic hydrocarbon at high temperature [59,60]. In this view, the dissociation of C-H bond of aromatic hydrocarbon drives reaction between carbon atoms and aromatic radicals that lead to the formation of crystallographic layered structure composed of hexagonal carbon ring [59]. Fundamentally carbon black is described in couple of terms i.e. carbon black aggregates and carbon black particles. The former term is coined for illustrating a discrete, rigid, colloidal entity composed of extensive coalesced particle while later term is exploited to ascertain small spheroidally shaped, aracrystalline, non-discrete components of an aggregates [60]. Carbon black in polymers are mainly exploited as conductive filler, dimensional and ultra violet light stabilizer. The incorporation of conductive filler like CB is supposed to form three-dimensional network in insulating polymer to impart the path of conductivity which is in turn defined as percolation or percolation threshold [61,62]. Kirkpatrick [63] and Zallen [64] have demonstrated that minimum 16% of volume fraction is required to develop three-dimensional network if spherical particles are distributed statistically on regular arrays. But Adriaanse et al. reported that the percolation threshold in CB based composite can be gleaned at very low volume fraction i.e. 0.03 vol% [65]. Gubbles et al. have published series of articles on design of conductivity in CB filled

polymer blends [66–68]. They have elucidated that selective localization of CB in multiphase polymer materials (in one phase or at the interface of binary polymer blend) possesses ability to reduce percolation threshold by many folds [67]. It is demonstrated that when CB reinforced in polystyrene (PS) matrix the obtained threshold is 8 wt% but in polyethylene (PE) and PS polymer blend at the composition of 45/55 respectively, selective distribution of CB in continuous PE phase further reduces the CB percolation by 5/3 factor. The selective distribution of CB at the interface of PE/PS for same composition (45/55) further diminishes the percolation to 0.4 wt% i.e. 0.002 vol%. Authors have reported that polymer/polymer interfacial tension and CB/polymer interfacial tension are essentially two decisive criteria for controlled or selective localization of CB in polymer matrix. The effect of CB content in epoxy matrix for antistatic application has been evaluated by Aal et al. where they have manifested that when weight fraction of CB in epoxy matrix is ≤ 15 wt%, the surface resistivity was observed in the range of 1.62×10^7 – $1.1 \times 10^4 \Omega \cdot \text{cm}$ [69]. When CB content further augment more than 15 wt%, the incorporated CB acts as electric conductor rather than a path for electric energy dissipation. In addition to conductivity, authors have also determined static decay time constant which exhibits the analogous results i.e. high decay time constant for lower CB content. The authors have demonstrated that the gap distance among the particles plays significant role in the final volume conductivity of the sample. It is reported that the increase in the filler concentration reduces the gap distance among the filler particles which in turn augment the crosslinking density results in increased composite conductivity. The authors have calculated the mobility carrier and density of the charge carrier to support their claims for high degree of bridging and intermixing of filler particle concentration. The equation of mobility carrier and the density of charge carrier has been elucidated as follows.

$$\mu = \left(\frac{v_0 e g^2}{KT} \right) \quad (4)$$

$$N = \frac{\sigma}{e\mu} \quad (5)$$

Where μ = mobility carrier; v_0 optical phenomenon frequency (normally $v_0 = 10^{-13}$ s); e = electron charge; K = Boltzmann constant; N = Density of charge carrier.

The low volume fraction of CB for developing conductive network in epoxy has been explained by Schueler et al. based on theory of colloidal particle which comprises of three features: particle-particle interaction, agglomeration dynamics and structural agglomerates [70]. They have elucidated that when a particle is dispersed in a liquid or polymeric medium, two competitive force are derived. One includes London-van der Waals attractive force between two neighbouring particles and second involves the

generation of repulsive coulombic force due to the charging of particle surface in the medium. The coulombic repulsive force constitutes potential barrier for particle to attract each other which can be engulfed by externally applied shear force or increasing the concentration of filler in the system. In the condition of externally applied load or absence of potential barrier, the neighbouring particle can form agglomerates which is eventually become more favourable in epoxy due to the low resin viscosity. Since circumstances are prevalent at various sites in the resin, agglomerates lead to the formation of conductive network even at low percolation thresholds. It has been demonstrated that the number of particles within such agglomerates can be represented as illustrated in Eq. (6) [70]. The small fractal dimension of agglomerates is essential to render the low percolation thresholds.

$$n = \frac{N}{\frac{4}{3}\pi R^2} a \frac{1}{R^{3-D}} \quad (6)$$

Where n = particle density; N = number of particles in an agglomerate; R = particle dimension; D = Fractal dimension.

The advent of carbon nanotube by S. Iijima [71] has stirred the shift of carbon fiber reinforced composite in avionic sector to the new class of nanocomposite based on carbon nanotubes [72]. Carbon nanotubes are mainly exploited in two predominant form single walled carbon nanotube (SWCNT) and multi walled carbon nanotube (MWCNT). SWNT can be simply visualized as rolled tube of graphite with the characteristics of metallic or semiconductor depending on the sheet direction about which the graphitic sheet is rolled [73]. The stated type of nanotube confides in the pair of integers (n, m) that drive the formation of armchair ($n = m$), zig-zag ($n = 0$ or $m = 0$) and chiral (any other n and m) [73]. SWNT demonstrates multiple eloquent properties including flexibility, ability to withstand cross-sectional and twisting distortion, extensibility in addition to the ability to withstand compression without fracture [74]. Though MWCNTs are formed by the arrays of concentrically nested SWNT but their properties cannot be explained by the physics of SWCNT. The relative stacking structure of MWCNT is persistent to the relative weak interaction between two adjacent layer which defines the position of atomic sites in outer layer relative to the position of inner layer of the tube [74]. The electronic structure of the MWCNT is considered as the sum of the constituent electronic structure of nanotube due to the their weak and incommensurate interaction [74]. The comparative properties of single walled carbon nanotube and multi walled carbon nanotube has been provided in Table 4 (reproduced with the permission of Mittal et al. [75] and Eder [76]).

The transition of polymer from insulator to conductor by incorporation of CNT can be obtained at low percolation threshold depending on type of polymer and synthesis methods, their aspect ratio, disentanglement of CNT agglomerates, degree of alignment and uniform spatial distribution of individual CNT [46]. Martin et al.

Table 4
Properties comparison between SWNT and MWCNT ^a.

Properties	SWNT	MWNT
Elastic Modulus/TPa	~1.4	~0.3–1
Strength/GPa	50–500	10–60
Specific gravity/(g·cm ⁻³)	0.8	1.8
Electrical conductivity/(S·cm ⁻¹)	10 ² –10 ⁶	10 ³ –10 ⁵
Electron Mobility/(cm ² ·(Vs) ⁻¹)	~10 ⁵	10 ⁴ –10 ⁵
Thermal conductivity/(W·(mK) ⁻¹)	6000	2000
Thermal stability/°C	~600	~600
Aspect ratio [77]	1000–10000 for percolation ≤ 0.1	1000–10000 for percolation ≤ 0.1

^a Reproduced with the permission of Elsevier from Mittal et al. A review on carbon nanotubes and graphene as fillers in reinforced polymer nanocomposites, Journal of Industrial and Engineering Chemistry [75] and American Chemical society from Dominik Eder, Chem. Rev. 2010, 110, 3, 1348–1385 [76].

Table 5
Electrical properties of CNT based polymer nanocomposite ^a.

Polymer type	Fillers	CNT/ wt%	Composite conductivity/(S·m ⁻¹)	Percolation threshold/(wt%/vol%)
Epoxy [85]	MWCNT	≤0.18	4 × 10 ⁻¹	-
Epoxy [86]	MWCNT	≤4	2 × 10 ⁻¹	0.5–1.0
Epoxy [87]	Pulmatic acid modified CNT	≤0.8	6.9 × 10 ⁻³	0.05–0.1
Epoxy [88]	MWCNT with and without non-ionic surfactants	≤12	5 × 10 ⁻⁵	0.072
Epoxy [89]	SWCNT	≤15	10	0.062
Epoxy [90]	MWCNT	≤0.5	0.01	≤0.1
Epoxy [91]	Oxidised MWCNT	≤1	10 ⁻²	0.012
PA [92]	SWCNT	≤13	10 ²	-
PA [93]	MWCNT	≈ 12	0.01	-
PU [94]	Modified MWCNT	≤7	10 ⁻¹	-
PU [95]	MWCNT	≤27	2 × 10 ³	0.009
PU [96]	MWCNT	3	30	-
Thermoplastic polyurethane [97]	MWCNT	1–5	-	-
Polystyrene/MWCNT microcapsule [98]	-	-	Surface conductivity (9.73 × 106 Ω)	0.5
isotactic polypropylene/poly(ethylene-co-1-octene) [99]	MWCNT	0–5 vol %	-	0.24 vol%
PU	MWCNT		Surface conductivity (7840; 78200; 26200 Ω)	0.2–0.5; 1–1.5; 0.6–1 depending on length of CNT

^a Reproduced with the permission of Elsevier from Spitalsky et al. Progress in Polym. Sci. 35, 2010, 357–401

have demonstrated the transformation of insulating epoxy matrix to bulk conductivity of 10⁻³ S/m by the reinforcement of aligned chemical vapour deposition (CVD) grown MWCNT [78]. The authors have mentioned that the aggregate's formation during composite processing entice significantly in the final conductivity of the polymer matrix which involves the utilization of high shear force and temperature. It is unambiguously explained that the high shear rate and high shear force (before the addition of hardener) endeavours the separation of nanotubes and subsequently small shear force or high temperature (after the addition of hardener) is required to overcome the mutual electrostatic repulsion of nanotubes [78]. In another study, the similar group has exploited the utilization of AC and DC electric field to align the MWCNT in epoxy composite and further demonstrated that the bulk conductivity of the composite enhances with the increasing strength of the applied field [79]. It is contemplated that AC field induces homogeneous nanotube network structure in polymer matrix compared to DC field of analogous strength therefore the obtained conductive in AC field is comparatively higher. The authors have theoretically demonstrated the attribute of carbon nanotube under the varying electric field and elucidated that the behaviour of nanotube in such condition is complicated due to the presence of varying conductance of each shell, available defects and further property difference by the virtue of varying size. They have contemplated that there are two types of contributing components of polarisation possible i.e. parallel to the tube axis and in the radial direction when electric field is applied in carbon nanotube. The presence of polarisation leads to the development of torque on nanotube which further aligns them against the viscous drag available in the surrounding medium in the direction of electric field. Such phenomenon is also dependent on the type of electric field i.e. DC or AC. Under the application of DC the charged CNTs move as per their electrophoretic mobility while in AC field the net electrophoretic mobility turned into zero by the virtue of zeta potential.

Sandler et al. in their study depicted the utilization of aligned CVD grown multi walled carbon nanotube in epoxy matrix with the varying weight fraction ranging from 0.001 wt% to 1 wt% with the percolation threshold of 0.0025 wt% (as per the percolation scaling law) [80]. They have elucidated the specific conductivity as a function of frequency where sample with 0.001 wt% exhibited increase in capacitive component with the frequency as followed in

pristine epoxy matrix. At the percolation threshold, frequency independent conductivity was observed below 10 Hz of frequency which further segmented with frequency increase while the conductivity become frequency independent with the further increase in weight fraction. The conductive characteristics of epoxy is further exemplified by Moisala et al. by incorporating multi walled carbon nanotube and single walled carbon nanotube (Chemically treated and ball milled) in the range of 0.005 wt% - 0.5 wt% [81]. They have reported that the percolation threshold in the case of MWNT was obtained at 0.0025 wt % while it was achieved at 0.05 wt% and 0.23 wt% for chemically treated (ultrasonicated) SWCNT in an ethanol solution saturated with sodium hydroxide) SWCNT and ball milled SWNT respectively. Apart from epoxy, various other polymer has been also exploited for the development of conductive nanocomposite based on CNT embodiments either in the form of SWCNT and MWCNT [82–84] as illustrated in Table 5 [84].

As evident from the Table 5, the electrical conductivity and percolation threshold of CNT filled polymer renders wide range discrepancy depending on their varying weight fraction in matrix which cannot be explained efficiently based on volume fraction [100]. In this context, contact resistance and intrinsic electrical conductivity (order of 10²–10⁵ S/cm in longitudinal direction [101] and 10⁰ S/cm in the transverse direction [102]) exhibits significant role in developing the conductive polymer composite [100]. Fiu-schau et al. have demonstrated that contact resistance or constriction resistance of two spherical conductive particles can be evaluated by Eq. (7).

$$R_{cr} = \frac{\rho_i}{d} \quad (7)$$

Where R_{cr} = constriction resistance; ρ_i = intrinsic filler resistance; d = diameter of contact spot.

It is essential to mention here that when the ratio of particle diameter to the diameter of contact in polymer matrix exceeds to 10, the above explained eq diverge. In the view of CNT the contact resistance for metallic-metallic contact and semiconductor-semiconductor is evaluated in the range of 100–400 kΩ [103] while this resistance is observed to be higher (≈ 3.4 MΩ) [104] for metallic-semiconductor contact. Grady has depicted that constriction resistance is essentially important at higher loading of CNT

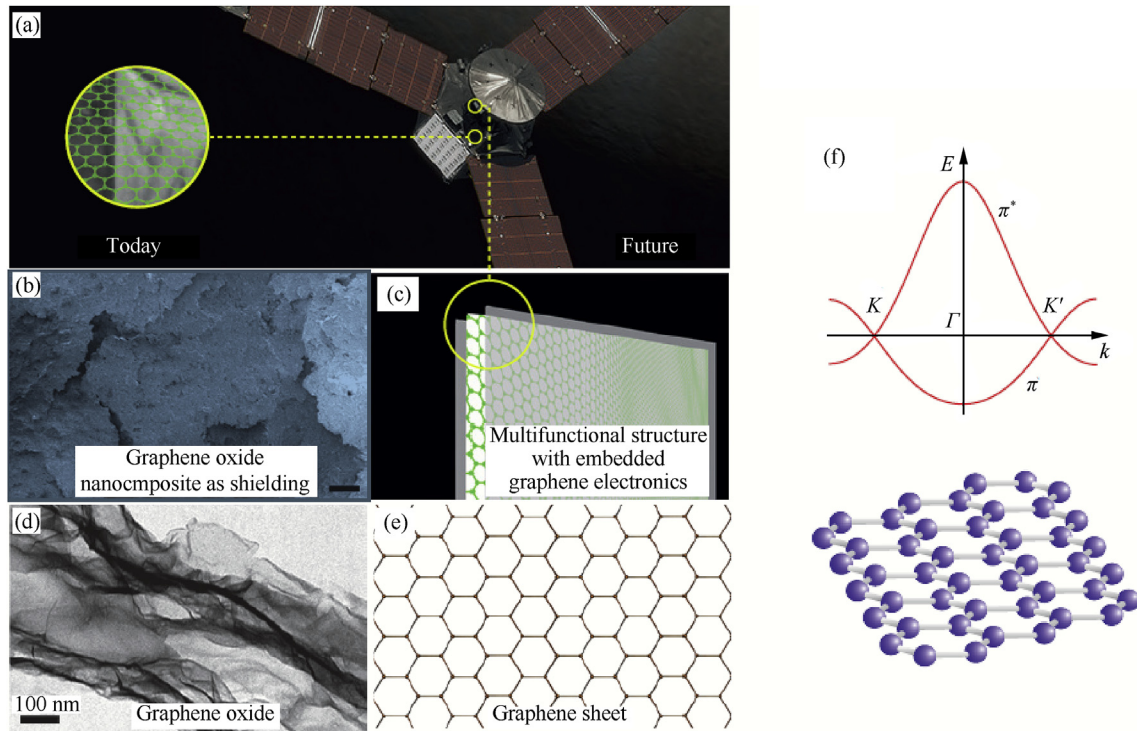


Fig. 4. Notional applications of graphene in spacecraft.

while tunnelling turned to be the prevailing in low volume fraction [105]. The tunnelling resistance evolve due to the formation of polymer film around the reinforced filler which considered as quantum mechanical process where charge transport transpires between conductive fillers without passing through the polymers [105]. It is considered that the phenomenon of tunnelling endeavours in polymer composite when the distance between adjacent particles are in the range of 5 nm or less. Li et al. have calculated this distance around 1.8 nm by employing Simmons formula and concluded that the contact resistance is dominating phenomenon compared to the intrinsic resistance of CNTs in CNT mats [106]. The complex conductive phenomenon, agglomeration and high of CNT in polymer matrix curtails the exploration of achieving its theoretical electrical and mechanical properties in real time composites [107,108]. Such detrimental characteristics of CNT trigger the utilization of CNTs in the form of sheets or buckypaper [109], fiber [110] and vertical arrays [111] but these forms are mainly exploited for high strength application aerospace as demonstrated by Siochi and Harrison [72]. In addition to CNT and other carbon-based fillers [112], Siochi has elucidated that graphene is also a futuristic promising materials to be exploited for aerospace application. He has provided notional application of graphene in aerospace application as depicted in Figs. 4(a)–(e) and concluded that though highly crystalline and single layer graphene is currently possesses constraints for utilizing as active conductive materials, but functionalized graphene can be exploited for electrostatic discharge filler with polymeric materials [113]. Graphene is a 2D hexagonal honeycomb lattice structure with sp^2 hybridization with string carbon bonds [114]. It is contemplated that each lattice of graphene encompasses 3 s bands and p -orbitals for all the carbon atoms are found to be perpendicular to the sp^2 hybridization plane. In this context, graphene achieves its excellent conductivity, high carrier rate ($15000\text{ cm}^2\cdot(\text{Vs})^{-1}$) at room temperature by the virtue of the movement of π electron in the plane. Graphene consists of two equal atoms that renders two equal tapered intersection point

(K and K') in each Brillouin zone, as a result undoped graphene elucidate zero band gap semiconductor (Fig. 4(f)) [115–117]. The unique characteristics of graphene entice the possibility of achieving electrical conductivity as high as 10^6 S/m [118] that enormously triggers its utilization in advance multifunctional composite [119]. In this context, Liang et al. demonstrated the utilization of graphene for the development of epoxy-based composite where they have reported percolation threshold as low as 0.52 vol% with the maximum conductivity of 0.05 S/cm for the approximately 8.8 vol% of loading [120]. In another study, Stankovich reported the fabrication of graphene based polystyrene composite with the percolation threshold as low as ~0.1 vol % with the conductivity of ~0. S/m at the reinforcement concentration of 1 vol% [121]. They have manifested that hydrophilic characteristics of graphite oxide can be reduced by its treatment with isocyanate that results in the formation of amide and carbamate ester bonds to the carboxyl and hydroxyl groups of graphite oxide. It is indicated that isocyanate

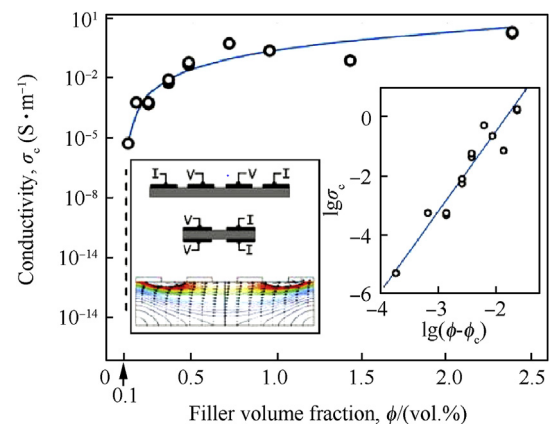


Fig. 5. Electrical conductivity of the polystyrene–graphene composites as a function of filler volume fraction.

Table 6Comparison of various lightning (LSP) strike protection systems ^a.

Type of LSP	Option	Main characteristics
Metallic mesh or foil bonded with resin	Mesh materials (copper, aluminum, bronze, nickel) Resin materials (epoxy, vinylester, modified epoxy) Prepreg materials or lay-up Foil, perforated or woven	High conductivity of metal Heavy surface material Problem with porosity
Metal or metalized fibers bonded with resin	Fiber materials: carbon, graphite, glass, polyester, synthetic fibers Coatings (nickel, copper, silver, platinum) PVD, SPS, Electroless, Thermal	Lightweight Less efficient than mesh Flexible process (multi coating and Multi-layer is possible)
Advanced solutions: polymer-based film or Conductive adhesives	Enhanced polymer with additives CNTs, Graphene sheets, Mix of CNTs and GS	Lightweight, Smooth finish, Must be replaced if struck

^a Reproduced with the permission of Elsevier from Gagné M, Theriault D. Lightning strike protection of composites. Progress in Aerospace Sciences. 2014; 64:1-16 [122].

treated graphite oxide renders exfoliated functionalized individual graphene oxide sheet thickness ~1 nm with the stable dispersant in aprotic solvents which can be further utilized for fabricating highly conductive polymer composite at low fraction of loading as illustrated in Fig. 5 (Reproduced with the permission of Springer Nature from Stankovich S, Dikin DA, Dommett GH, Kohlhaas KM, Zimney EJ, Stach EA et al. Graphene-based composite materials. Nature. 2006; 442:282 [121]).

Apart from various nanofiller reinforcement, Gagné and Theriault have provided detailed description over various approaches to manipulate the structure of composite or composite itself against the lightning strike and static charge accumulation [122]. They have elucidated that metallic mesh and metalized fiber bonded with resins of epoxy, vinyl ester or modified epoxy are another class of materials which can be exploited for the development of composite for electrostatic discharge application in aircraft. They have tabulated the various protection system and their characteristics as described in Table 6.

In Fig. 4, (a) Different parts of a spacecraft could host different types of application; (b) graphene oxide nanocomposite (Scale bar-40 μm); (c) multifunctional structure with embedded graphene electronics; (d) graphene oxide; (e) graphene oxide sheet. Fig. 4(a–e) reproduced with the permission of Springer Nature from Siochi EJ. Graphene in the sky and beyond. Nature Nanotechnology. 2014; 9:745 [113]. (f) Schematic representation of crystal and electronic band structures (only π -bands are shown) of free-standing graphene. (Reproduced with the permission of Springer Nature from Novoselov K. Graphene: Mind the gap. Nat Mater. 2007; 6:720 [115]).

Onion like carbon is another class of carbon material utilized for imparting conductivity of the polymer materials in the recent years. D. Ugarte have reported that OLC possesses quasi-spherical graphitic system consisting concentric arrangement of nearly perfect sphere with inner most spherical size of typical C_{60} (0.7–1 nm) [123]. Such carbon structure can be generated by high-energy electron irradiation, shock wave treatment of carbon soot, carbon deposits in plasma torch and laser melting of carbon etc. Kuznetsov et al. have demonstrated the formation of OLC by heat treatment of ultra-dispersed diamond powder of size 2–6 nm [124]. Zeiger et al. have recently provided the detailed review on nano-diamond derived onion like carbon and discussed the electrochemical properties of different types of carbon onions as electrode materials [125]. One example of transforming nano-diamond into onion like structure can be obtained in Fig. 6 (Reproduced with the permission of Royal Society of Chemistry from Zeiger M, Jäckel N, Mochalin VN, Presser V. carbon onions for electrochemical energy storage. Journal of Materials Chemistry A. 2016; 4:3172–96 [125]). E. Palaimiene et al. have exploited the utilization of onion like carbon (OLC) for the development of epoxy based conductive composite [126]. They have demonstrated that, when the size of the OLC

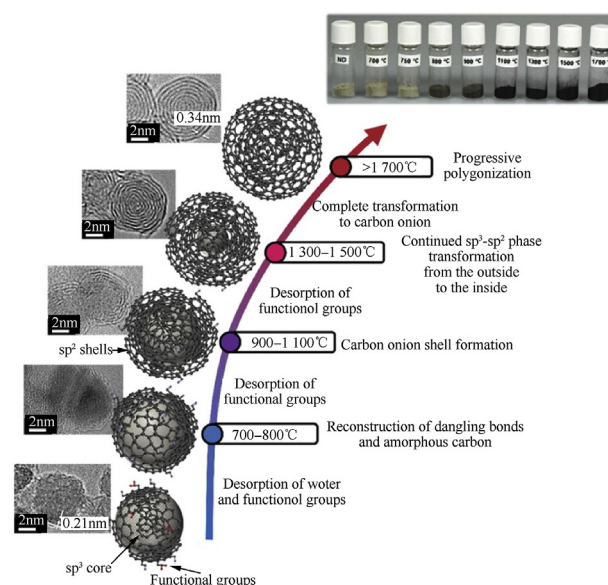


Fig. 6. Transformation from nanodiamonds to carbon onions by annealing shown using transmission electron micrographs, optical images, schematic illustrations of intermediate steps, and the assignment of physical effects depending on the annealing temperature (slight variations occur depending on the annealing atmosphere, especially when comparing vacuum with an inert gas atmosphere).

particles increase combined with shear stress in epoxy matrix, resulted in the formation of extended pearl like structure by the virtue of opening their loose packed structure. They have also reported that the bigger size of OLC aggregates (250 nm) possesses ability to render the lowest percolation threshold (0.7 vol%) for conductivity.

5. Conclusions

The review enunciated the progress in reducing or controlling the accumulation of statics that could cause disasters depending on the threshold of accumulation and the environment subjected too. The statics has been a threat to aviation flight safety leading to either blinding the aircraft or making the communication blank momentarily and severe effects of explosion when interacting with explosive medium. This has further aggravated with composites substituting metals in aviation structures and avionic parts. The dielectric surfaces are more prone to static accumulation which has led to the puncture of surfaces when struck by lightning leaders. The remedial measures in form of anti-static materials for composites have paved their way since 19th century with the introduction of nanomaterials. The various matrix-filler combinations

have been discussed to suit the applications in aviation where epoxy was conceived as the most common matrix with the reinforcement of carbon-based nanoparticles like carbon black, carbon nanotube (multi walled and single walled), graphene etc. The higher loading or non-uniform dispersion challenges has led to agglomerations and deterioration in electrical and structural properties. Thereby, further research is ongoing in determining suitable replacements such as the carbon nanofibers and other techniques to surmount the deficiencies related to poor dispersion and agglomerations in conductive composites.

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