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A Meta-Study on Smart Coatings with Carbon Nanoparticles

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Abstract

Smart coatings that are rooted on polymer matrices are usually doped with carbon nanoparticles such as graphene or carbon nano-tubes and are being widely researched. Information was gathered from over 74 articles that focused on the smart coatings of polymer doped with carbon nano-particles databases such as ResearchGate, Academia, PubMed, Scopus, Web of Science, Google Scholar, ScienceDirect, and IEEE Xplore. Articles were filtered by keywords including 'smart coatings', 'polymer nano-composite', 'carbon nano-particles', 'deicing', and 'self-heating' to identify scientific articles relevant to this present research on smart coatings and carbon nano-particles. From the meta-study, it was revealed that in organic coatings, carbon nano-fillers frequently increase performance by improving corrosion resistance, barrier properties, hardness, and wear strength. Additionally, the identified composites represent a new class of protective organic coatings capable of autonomously responding to environmental stimuli or damage. Through the investigation, it was understood that polymer coatings are new functionalities that are added by carbon nano-particles and, as a result of the formation of a percolation network, the majority of which are connected to the nanocomposites, increased electrical conductivity. Based on the fluctuation of their electrical resistance, these coatings can be employed as strain sensors and gauges (Structural Health Monitoring (SHM)). Additionally, they perform the task of self-heaters by applying electrical power connected with resistive heating via the Joule effect. This brings up new opportunities, notably for deicing and defogging coatings. The lotus effect is used to develop micro- and nano-scaled hierarchical surfaces for superhydrophobic and self-cleaning coatings. Surface damage can be repaired by coatings having a self-healing polymer matrix.

Keywords: Smart coatings, Polymer nano-composite, Carbon nano-particles, Deicing, Self-heating.

1 | Introduction

Smart coatings are unique covering materials that can detect and respond to environmental stimuli. They are constructed from programmable materials that react to changes in light, chemical, thermal, or other stimuli.

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Because of their piezoelectric, thermoelectric, piezo-resistive, and chemical qualities, they gain additional capabilities, such as self-sensors, self-cleaning, and self-healing. The majority of today's smart coatings are made from nano-reinforced polymers. The addition of inorganic and functional organic nano-fillers to polymers often enhances their thermal and mechanical characteristics while also adding new functionality. As it is widely acknowledged, one of the primary benefits of using nano-fillers is their high specific area, which decreases the nano-filler content greatly and improves load transfer from the matrix when the interface is adequate.

The focus of this study was on the inclusion of carbon nanoparticles in this work, namely Carbon Nanotubes (CNT), Graphene Nanoplatelets (GNPs), and Graphene (G) [1], [2]. They have a unique combination of mechanical properties, outstanding thermal and electrical conductivity, and high toughness and stiffness [3]. They are non-toxic and have a little carbon footprint [4]. They are classified as multifunctional polymer matrix fillers for all of these reasons. Indeed, polymer nano-composites reinforced with carbon nano-particles frequently demonstrate better electrical, thermal, and mechanical properties, as well as innovative smart material performance [5]. Because of their piezoresistive property, they can be used as strain sensors by changing the electrical resistance of the composite as a result of deformation of the electrical network caused by graphitic nano-fillers. Nano-fillers, on the other hand, can be used as actuators, such as self-heaters or chemical absorbents due to Joule heating. The matrix is a sophisticated stimulus-responsive polymer in this case, and the carbon nano-fillers are the stimuli that trigger the polymer to respond.

2 | Methods

The meta-study draws on literature focusing on the smart coatings of polymer doped with carbon nanoparticle databases such as ResearchGate, Academia, PubMed, Scopus, Web of Science, Google Scholar, ScienceDirect, and IEEE Xplore. At first, the research was directed towards smart coatings with respect to carbon nanoparticles to achieve a broad understanding of the topic. The essence was to identify pertinent data and insight to fine-tune the broad subject into two focal points. There has been an emphasis on carbon nanoparticles since 2003, ensuring the relevance of the data collected due to rapid advancements in Technology.

The keywords which helped obtain suitable data included ‘smart coatings’, ‘polymer nano-composite’, ‘carbon nano-particles’, ‘deicing’, and ‘self-heating’ to identify scientific articles relevant to this present research on smart coatings and carbon nanoparticles. The sources employed for this meta-study were from the journals and scientific articles earlier mentioned particularly as independent sources which provide more relevance. Scientific journals concerning the organic coatings, carbon nano-fillers, and the representation of composites were investigated.

3 | Results

3.1 | Descriptive Analysis of Articles

For the entire research, information was gathered from 74 articles that focused on the smart coatings of polymer doped with carbon nano-particles from databases such as ResearchGate, Academia, PubMed, Scopus, Web of Science, Google Scholar, ScienceDirect, and IEEE Xplore. The period for 2003-2020 and the deadline for retrieving the articles was 10th May 2022. The highest number of articles retrieved (10) were published in 2018, followed by 9 articles in 2016 and 2017, respectively (see *Fig. 1*).

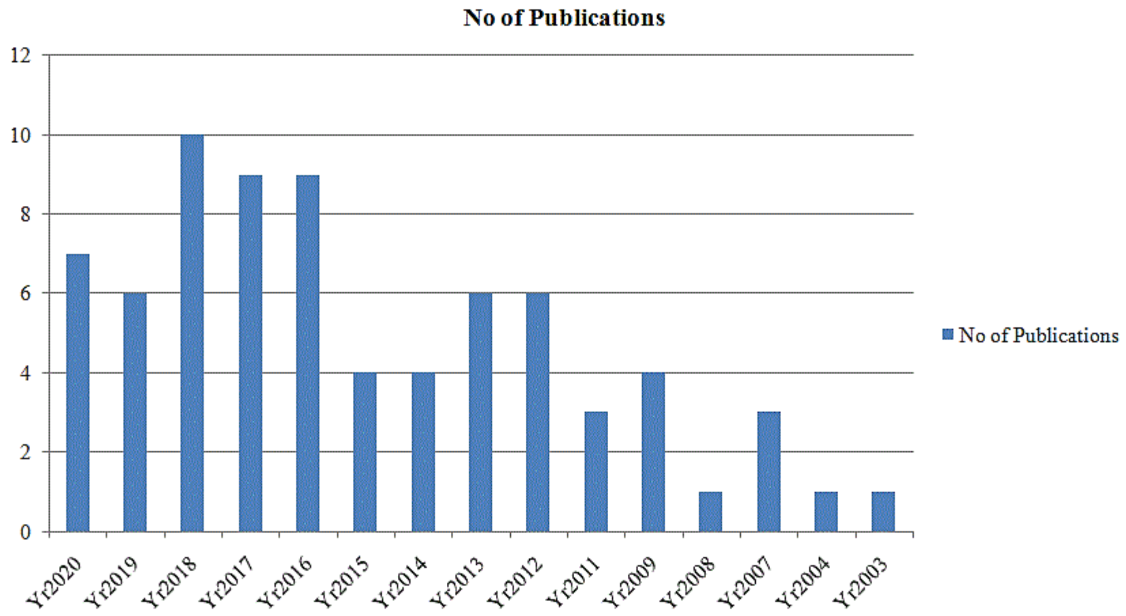

Fig. 1. Total number of publications.

Regarding the processing and synthesis of nano-composite polymer coatings and the characteristics of nano-composite polymer coatings, information was gathered from 15 articles. Regarding the smart nano-composite coatings, information was gathered from 23 articles. Regarding the fundamentals of self-heating by the Joule effect and self-heating of smart nano-composite coatings, information was retrieved from 8 articles. Regarding the deicing system in the form of self-healing, information was gathered from 7 articles. Regarding the self-curing coatings, information was gathered from 3 articles. Regarding the self-healing coatings, information was gathered from 4 articles. Regarding self-cleaning using smart nano-composite coatings, information was gathered from 6 articles.

3.2 | Processing and Synthesis of Nano-Composite Polymer Coatings

Processing and synthesis of nano-composite polymer coatings typically involves at least two distinct stages:

I. Dispersion of nano-fillers into monomers, prepolymers, or polymers.

II. Coating manufacture.

A variety of methods may be used to distribute carbon nano-filler into monomers and polymers. In reality, multiple articles and reviews have been published. As a result, they are just briefly described in this section. The improvement of properties on nano-composites, particularly mechanical properties, is well known to be strongly dependent on dispersion quality, which is rooted in the physical and chemical interaction between functionalized nano-fillers and the polymer matrix. Excellent nano-particle dispersion is essential to develop a high-performance nano-composite.

Direct mixing, in situ polymerization in the presence of nanoparticles, and solution mixing are the three most common nanocomposite processing methods. In situ particle synthesis is another way of processing that is based on the in situ sol-gel process within polymers, although it is only used for inorganic nano-fillers. Carbon nano-fillers must be applied to polymer or prepolymer surfaces, depending on the polymer type. During the polymer production process, such as calendaring or extrusion, nano-filler dispersion on thermoplastic polymers is routinely performed. Nonetheless, prior to curing into prepolymers or monomers, nano-filler dispersion on thermosetting and rubber polymers is commonly undertaken.

Different dispersion techniques, depending on the application of mechanical forces or an electric or magnetic field, can also be used in this final situation. As previously stated, the second phase is creating one's coating by utilizing several conventional coating processing processes. Cold spray processing is often utilized for polymer nanocomposite coatings to minimize heat degradation of the substrate. Other applicable manufacturing procedures include latex, emulsion, and dispersion in situ polymerizations.

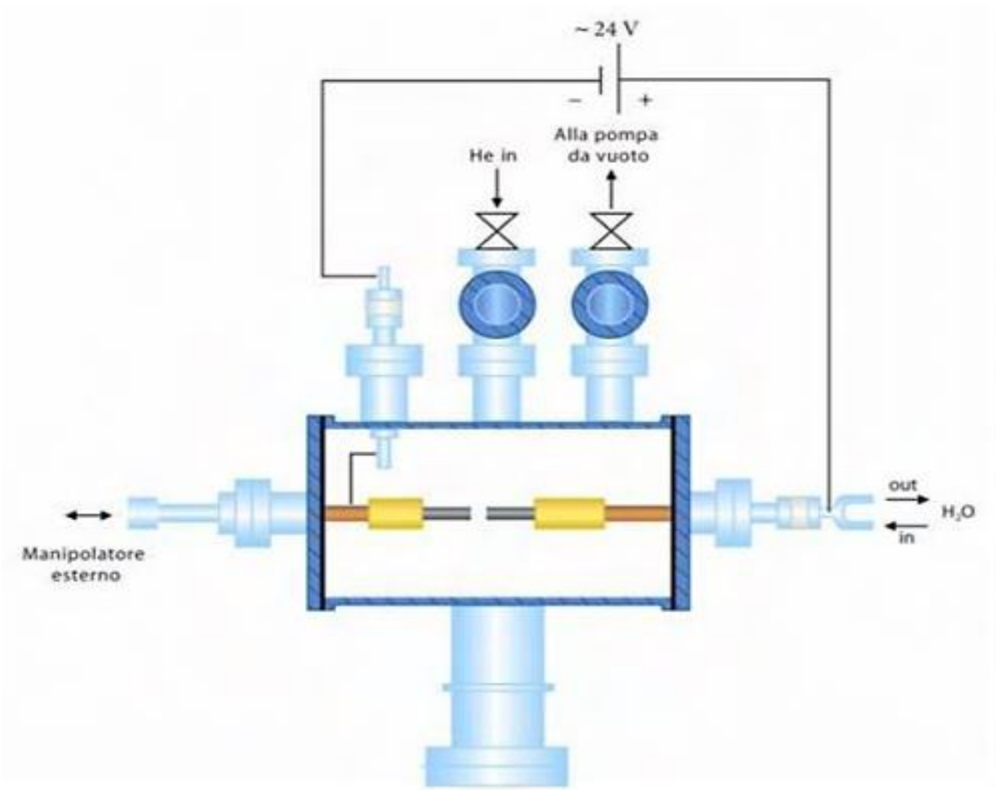

Fig. 2. Design of arc discharge synthesis chamber.

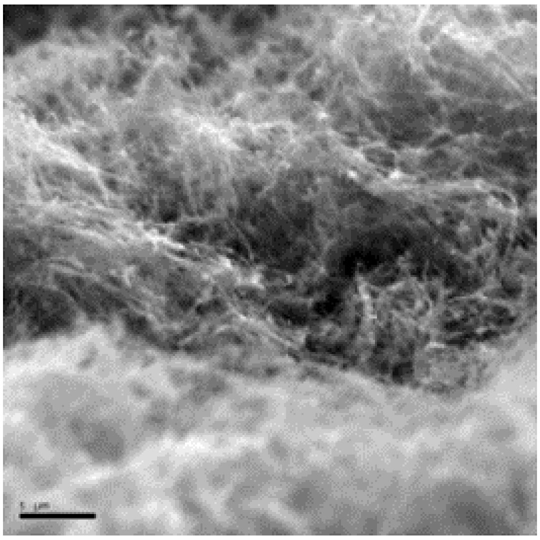

Fig. 3. S.E.M image of arc discharge carbon nanotubes.

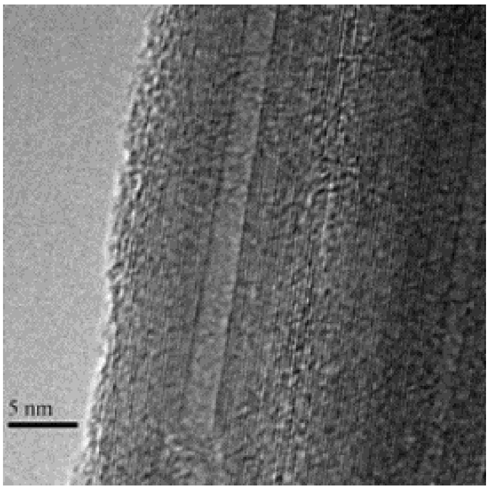

Fig. 4. T.E.M. Image of Arc discharge Carbon Nanotubes.

3.3 | Characteristics of Nano-Composite Polymer Coatings

To increase the mechanical characteristics of polymer coatings, graphitic nano-fillers are often utilized. Because graphite is a solid lubricant, graphitic nano-fillers can be used to improve the tribological performance of polymer coatings. It is feasible to create graphene-containing polymer coverings with good tribological characteristics, such as low wear rate and low friction coefficient [5], [6]. The addition of graphene gradually reduces the wear rate and friction coefficient of the composite coating. Graphene-reinforced thermosetting coatings outperform plain coatings in terms of wear resistance and friction reduction at high temperatures. The value of these properties increases as graphene content increases [6].

The types of composite nanocomposite are shown in *Figs. 4-6*, respectively. It is worth noting that melt intercalation is known to be the most common method for creating thermoplastic polymer nanocomposites. As shown in Fig. 8, it entails annealing the polymer matrix at high temperatures, adding the filler, and, lastly, kneading the composite to ensure uniform distribution. It offers the benefit of being environmentally friendly due to the absence of solvent consumption. Furthermore, it is thought to be compatible with industrial processes like injection molding and extrusion, making it easier to use and hence more cost-effective. Nonetheless, the high temperatures utilized in the technique might harm the filler's surface modification.

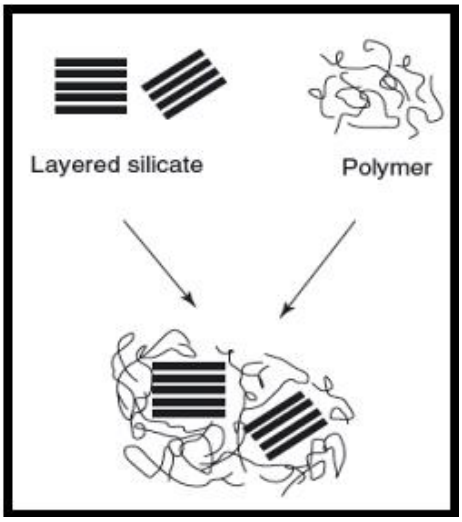

Fig. 5. Unintercalated nanocomposite.

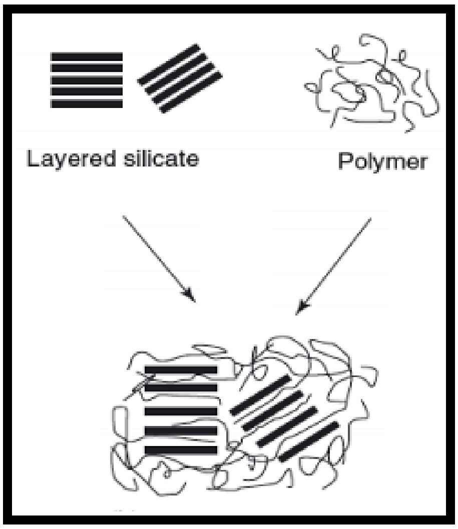

Fig. 6. Intercalated nanocomposite.

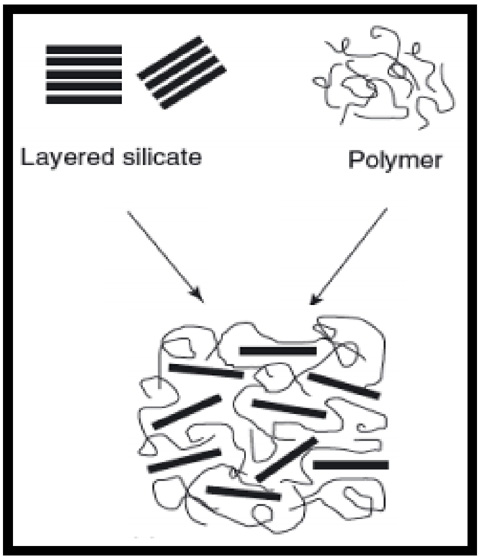

Fig. 7. Exfoliated nanocomposite.

Meanwhile, the friction coefficient and wear rate of graphene/composite coatings exhibit no obvious trend as temperature rises. The creation of a surface transfer layer, which dampens the high contact pressure and heat, might be connected to this phenomenon [6], [7]. Self-lubrication, wear-proof, and anti-friction (CNT/polymer coatings) are properties that can enhance wear resistance, particularly friction [7], [8]. However, a number of factors influence their tribological behavior, such as the composition and properties of sliding pairs (surface roughness and main mechanical properties like hardness, stiffness, and fracture toughness), as well as sliding parameters such as speed, load, lubrication state, and temperature among others.

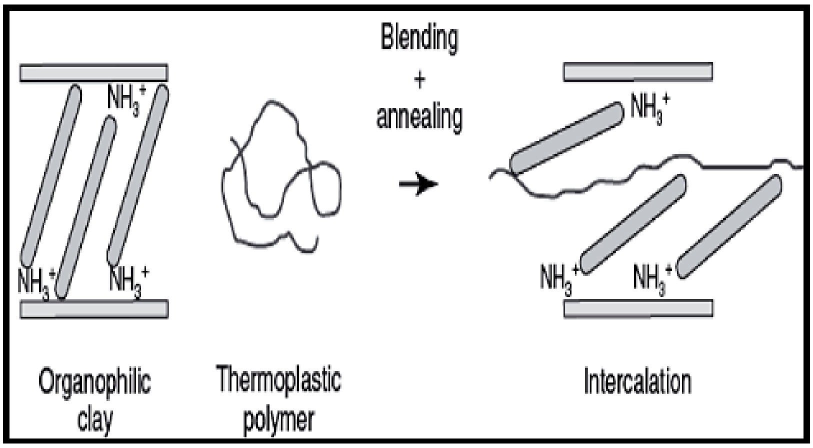

Fig. 8. The melt intercalation process.

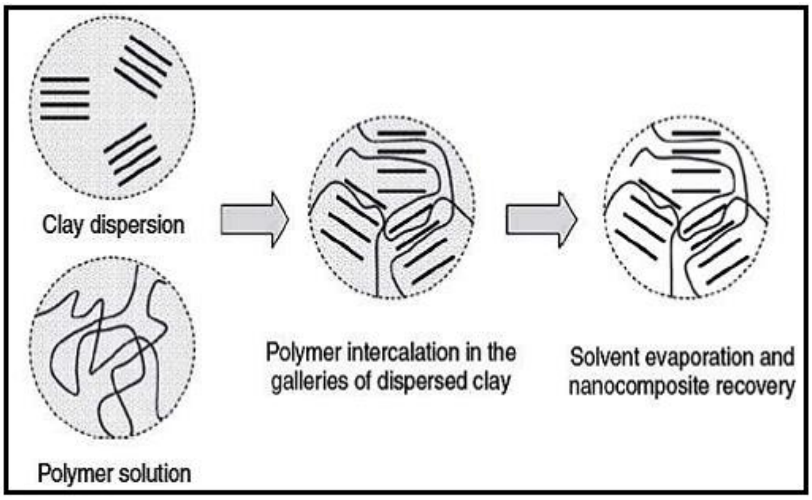

Fig. 9. The exfoliation adsorption process.

This behavior is characterized by the following processes: bridge crack of CNT and crack propagation lock, lubricating action via dislodgement of individual graphene layers, strengthening of reinforced polymer matrix and heat dissipation, and temperature-driven wear reduction [6–8]. It is worth noting that the optimal carbon nano-particle content for achieving the best tribological characteristics exists. Nonetheless, this amount is determined by a variety of parameters, including the aspect ratio of the nano-filler, the degree of dispersion, interaction, and readings of the nano-fillers with the polymer matrix at the edges [7]. The inclusion of carbon nanoparticles in polymer composites enhances their durability. The hardness of the composite coating improves as the quantity of graphitic nano-filler grows; however, the slope of the curve decreases as the amount of graphitic nano-filler increases due to agglomerations in the composite coating [9].

Anti-corrosive coatings are one of the most important uses of polymers augmented with graphitic nano-fillers. Anti-corrosive coatings utilize corrosion protection technologies such as cathodic protection, barrier protection, electrolytic inhibition, active corrosion inhibition, and anodic passivation [8]. The "nano-barrier wall effect" and "torturous path effect" of graphitic nano-particles improve polymer coating barrier properties, which are highly dependent on nano-orientation, filler dispersion and exfoliation degree, polymer-nano-filler interface, aspect ratio, and crystallinity of cross-linking degree of thermosetting resins or thermoplastic polymer [8]. The presence of nano-fillers causes twisting routes, which lowers the diffusion coefficient.

The zigzag diffusion channel provided by graphene's orientation and vast surface area inhibits corrosion species from spreading. Furthermore, because of their high electrical conductivity, electrons are prevented from exiting the cathodic site by providing an alternative path [10]. The graphitic nano-fillers embedded with polar groups enhance the ionic resistance of the coating by creating a negative charge on the facade of the graphitic nano-fillers, thereby averting the movement or flow of hydroxyl anions and chlorine when exposed to neutral and alkaline environments. Super-hydrophobic surface development has garnered significant attention in recent studies [11]. Agricultural, textile, vehicle construction, marine, and optical sectors all utilize super-hydrophobic coatings due to their remarkable water-repellent properties.

Three components are necessary to create a super-hydrophobic surface: nano- and micro-scaled hierarchical surfaces, micro-scaled roughness, and low surface energy [12]. The hierarchical multi-scale surface in coatings with a high CNT content may be generated by creating CNT agglomerates at the micro-scale and CNTs themselves at the nano-scale. Polyvinylidene Fluoride (PVDF) is a popular hydrophobic polymer coating [13]. The addition of GNP results in a comparable rise in hydrophobicity [14]. Super-hydrophobicity is commonly requested in conjunction with additional qualities like self-cleaning, anti-icing, and deicing, which will be covered in further detail below.

The inclusion of graphene into polymers improves polymer composite thermal stability. The larger specific area of carbon nano-fillers enhances more stability [13]. This improvement is also accompanied by a significant rise in polymer thermal conductivity. Extremely high improvements have been recorded, ranging from 0.2 W/mK for a clean epoxy coating to 20 W/mK for a composite containing 30% graphene. Graphene is a more effective nano-filler than CNT in improving the thermal conductivity of polymer composites. Graphene's thermal conductivity is ascribed to phonons and electrons, which justifies its high thermal conductivity (2000 W/mK). Copper, for example, is an excellent thermal conductor with a conductivity of 400 W/mK. The contribution of phonons, in this case, is restricted to 1–2 percent of the aggregate.

Heat conductivity may be increased by 50% when non-functionalized graphene is used. When graphene is treated with functional groups capable of making covalent and non-covalent bonds with the polymer matrix, the increase can be as high as 100%. Furthermore, when graphene is coated with a titanium-coupling agent, heat conductivity increases by 300 percent. Researchers have explained this as a result of surface modification decreasing the interfacial thermal resistance between graphene and polymer [15]. For the same reason, a greater degree of dispersion of graphene inside the polymer matrix enhances the composite's heat conductivity. It is well understood that the presence of graphitic nano-fillers promotes electrical conductivity [16].

Nano-fillers with a higher specific area and aspect ratio significantly enhance electrical conductivity, achieving an improvement from 10-8 to 10-10S/m for separating polymer composites. In this case, the functionalization of nano-fillers usually results in a minor increase in composite electrical conductivity. The decrease in electrical conductivity of neat nano-fillers is attributed to the partial breakage of some C-C structures during functionalization, as well as the surrounding isolating polymer layer formed over the functionalized nano-fillers, which prevents direct contacts between electrical nano-particles [16].

To increase the Fire Resistance (FR) of combustible substrates, nano-reinforced coatings are being researched. Graphene decreases Total Heat Release (THR) in general by inhibiting the generation of dangerous gases during thermal breakdown due to its radical trapping and multilayer hindering effect. CNT additionally improve FR performance by reinforcing carbonized layers [16]. They also serve as a good physical barrier, greatly decreasing the Peak Heat Emission Rate (PHRR).

Synthesizing CNT granules (durobeads) with a polymer coating

To disperse the CNTs, a homogenizer at 6,000 rpm was used to mix K-Nanos 100P (45 g) of 7–23 nm diameter with clean water (2.5 L), as shown in *Table 1*. The dispersion can be transported to a container equipped with a screw mixer of 1,000 rpm, and 360 ml of toluene, which contains olefin-based melted polymer with a melting temperature of 103oC will be added to form spherical granules which range from 0.5 to 2.0 mm, diameter. The solution will be finally prepared by filtering it through a 60-mesh metal screen and drying it for 7 hours in a vacuum oven set to 70oC, as shown in *Figs. 10* and *11*. Granule formation, also known as "granulation", is feasible with 0% polymer in the synthetic system shown in *Fig. 12*. CNT granules were sonicated prior to use for in vitro and vivoexperiments.

Table 1. Description of K-Nanos 100P.

Item	K-Nanos 100P
Diameter	7–23 nm
Bundle length	26 nm (average)
Bulk density	0.015 g /ml
Carbon purity	~ 95%
Polymer %	**Bulk density (g/ml)**
K-Nanos 100P	0.015
0%	0.095
6%	0.126
10%	0.148
20%	0.148

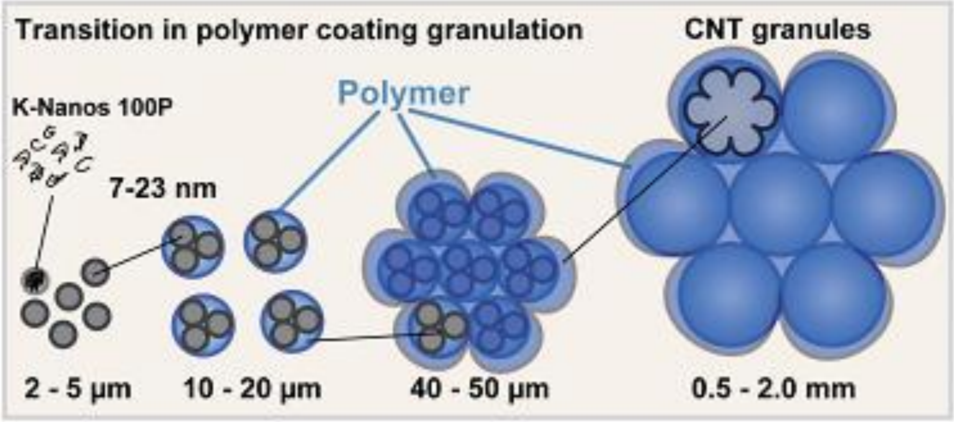

Fig. 10. Procedure to generate CNT granules with a polymer coating.

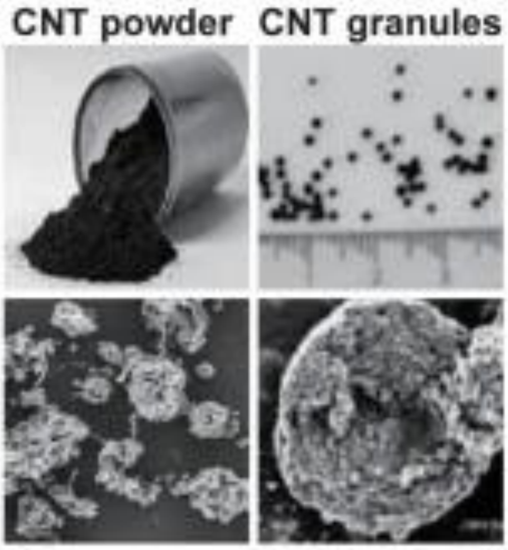

Fig. 11. Metamorphosis of K-nanos 100P from CNT powder to granules. Lower panels Scanning Electron Microscopy (SEM).

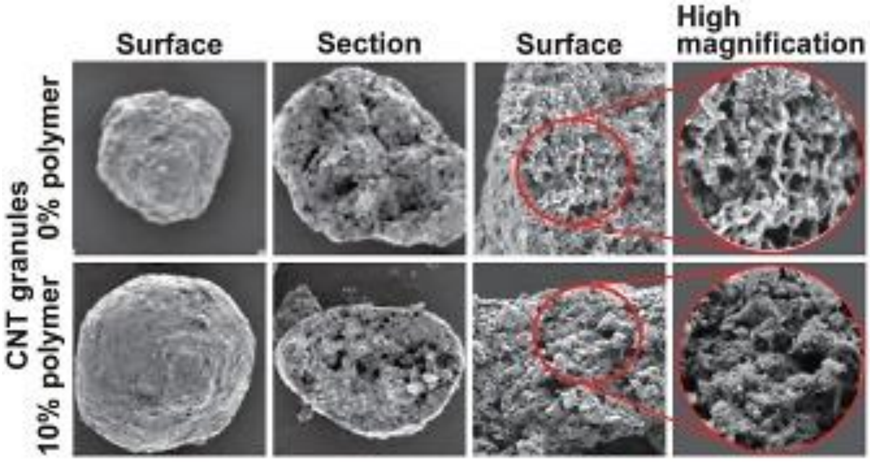

Fig. 12. SEM images in CNT granules with or without polymer coating.

3.4 | Smart Nano-Composite Coatings

The notion of Structural Health Monitoring (SHM) is of tremendous interest in polymer coatings due to its ability to provide an online evaluation of the extent of damage. A competent SHM approach in this context must achieve the four stages outlined by: detection, localization, damage measurement, and assessment of remaining life, commonly known as prognosis.

There are several SHM methods available now, including lamb waves, fiber optics, and acoustic emission, among others. However, these methods frequently involve complex mathematical and statistical techniques and do not always provide a comprehensive picture of the structure's health. Consequently, the development of more effective and holistic SHM approaches is currently receiving significant attention [17].

Basics of SHM embedded with carbon nano-particles

As previously stated, carbon nano-particles have distinct mechanical and, notably, electrical characteristics when compared to other materials. As a result, their incorporation into an insulating medium stimulates the formation of electrical networks. This phenomenon causes a significant increase in electrical conductivity, making the polymer covering electrically conductive. First and foremost, the idea of percolation threshold must be defined. It is the crucial volume fraction of nano-particles in which an efficient electrical network, permitting current passage, is generated. Various aspects determine it, the most important of which are the geometry of the nano-filler (including its 0D, 1D, or 2D nature) and its dispersion state or their distribution within the nano-composite.

The percolation threshold is an important aspect in determining the minimal amount of nano-filler required for electrical applications. Furthermore, the impact of different nano-particle network characteristics on the percolation threshold has been extensively investigated in recent years. Li et al. [18] presented a simple analytical model that correlates geometry, aspect ratio, and dispersion state with percolation threshold value. They determined that the lower the aspect ratio and the greater the degree of agglomeration of nano-particles, the higher the percolation threshold. Because of their low Carbon Black (CB) aspect ratio, reinforced polymers have very high percolation threshold values.

Bauhofer et al. [19] examined the influence of nano-filler shape and dispersion method on percolation threshold in CNT-based polymer nanocomposites. It was discovered that the most vigorous dispersion techniques, such as ultrasonication, although resulting in the most homogenous distribution of nano-particles, result in considerable CNT breaking. This widespread aspect ratio lowering results in higher percolation threshold values. The impact of the dispersion method, however, differs depending on the nano-filler. In this example, ultrasonication proved to be an effective dispersion method for GNP-based nanocomposites. GNPs are made up of many layers of graphene and are commonly utilized as reinforcement in polymer nano-composites due to their inexpensive cost.

In this case, ultrasonication induces graphene layer exfoliation [18]. As a result of the increased aspect ratio of the nano-fillers, these exfoliation processes cause a lowering in the percolation threshold. Because of the permutation of exfoliation stretching impacts, the permutation of three roll milling and ultrasonication stages can permit the GNP nano-composites production withsignificant reduced percolation thresholds. As a result, the notion of SHM with nano-particles is based on the monitoring of changes in the electrical network when subjected to strain or damage. However, in order to have a better grasp of these SHM approaches, it is necessary first to comprehend the fundamental conducting processes in the electrical network.

Three independent mechanisms may be identified here: the nano-filler's intrinsic conductivity, contact between adjacent nano-particles, and tunneling transport between two neighboring particles that are not in close proximity. Tunneling transit is vital in the electrical network of a nano-composite. The reason for this is that the corresponding tunneling resistance is many orders of magnitude bigger than the intrinsic and contact resistance. As a result, when the electrical network is subjected to strain or damage, the variation in the tunneling distance between nano-particles governs the alterations.

Simmons [20] identified a linear-exponential connection between tunneling resistance and inter-particle distance, which is commonly referred to as tunneling distance. It demonstrates that the electrical resistance increases with increasing distance between neighboring nanoparticles. More specifically, when subjected to strain, the electrical resistance fluctuates, increasing the tunneling distance between neighboring nanoparticles.

Sensitivity of polymer-based nanocomposites

In this situation, the previously established percolation threshold concept is critical. In actuality, the linear-exponential connection suggests that the larger the tunneling distance in the initial condition, i.e., without the application of any strain, the increase in the resistance of electrical variation connected with the tunneling distance fluctuation when strain is applied. The Gauge Factor (GF) is defined in this context as the variation of the normalized resistance R=R0 divided by the applied strain. As a consequence, in order to acquire the highest GFs, it would be necessary to deal with nano-filler volume fractions around the percolation threshold, as the distance between neighboring nano-particles will be the biggest possible to create an efficient electrical network, and as a result, the range in tunneling distance will be the greatest.

This was identified in both CNT and GNP nano-composites, with concentrations close to the percolation threshold giving the largest GFs [21]. However, there are substantial variances between the various nano-particles in this regard. For example, GNP-based nano-composites display a more prominent exponential trend of electrical resistance with applied strain [21] than CNT-based nano-composites. It means that at low strain levels, GF values are substantially lower than at high strain levels. The many interactions that take place inside the electrical network can explain the alleged exponential behavior of GNP nano-composites. In practice, the tunneling area of these 2D particles is usually much bigger than that of CNTs, meaning that the inter-particle distance may be much greater for efficient tunneling transport.

As earlier stated, the stronger the exponential connection between electrical resistance and applied strain, the greater the tunneling distance. Furthermore, because there is a relationship between the exponential behavior and the sensitivity of the nano-composite, GNP-based nano-composites have higher GF values than CNT nano-composites (from 12–15 to 2–4 at low strain levels, respectively, for nano-composites manufactured using similar techniques) [21].

Adoption of SHM in nano-composite coatings

The enormous potential of the nano-particles for SHM applications has been widely exploited in the development of surface sensors and smart coatings. More precisely, their application as alternatives to strain gages has been receiving interest lately. A strain gauge is essentially a gadget that indicates the strain level of a structure at the place of connection. Too far, the most widely used are based on traditional metallic foils, with the strain acquired from the electrical resistance change caused by the deformation of the metallic foil when subjected to this strain level. The GF values of traditional strain gauges are typically in the range of and exhibit a relatively linear correlation of electrical resistance change with applied strain.

The creation of very flexible sensors is the primary goal of strain-sensing device research using carbon-based nano-composites. Carbon nanoparticles, CNT, and graphene have all gotten much attention in this sector. The inclusion of these nanoparticles into high-strain capacity polymers, such as fluoro-elastomers [22], Thermoplastic Polyurethane (TPU) [23], or vulcanized silicone [24], has shown outstanding sensing capabilities. Because the GF at high strain levels (>20 percent) may range between 400 and 4000 depending on the quantity and form of the carbon nano-filler, they have significant promise for wearable electronics or human motion sensing.

At higher strain levels, the carbon nano-particle network exhibits a highly prominent exponential behavior due to the abundance of tunneling mechanisms. Furthermore, they may be used as pressure sensors, with improved sensing capabilities when compared to others, since the strain-induced failure that they promote allows them to detect extremely minute pressure changes. When compared to bulk nano-composites, which are designed to have a 3D uniform placement of the nano-fillers, the 2D nature of the electrical network in a nano-composite coating supports a rise in the percolation threshold [25]. Furthermore, the cross-sectional area of the coatings is evidently considerably less than that of 3D nano-composites, implying that the electrical resistance is substantially larger.

As a result, the amount of nano-filler required for SHM purposes in polymer coatings is substantially larger. Furthermore, their SHM capabilities in various polymer coatings based on thermosetting polymers have been widely shown, in addition to flexible devices such as human motion sensors, wearable electronics, or strain gauges. GNP-based coatings, in particular, have demonstrated high sensitivity to compressive and tensile loading at low strain levels, as well as good reproducibility under cyclic loads. Furthermore, CNT-based ones have shown strong sensing characteristics with great linearity [26]. Although their interest in strain-sensing devices is high, their crack-sensing capabilities may be even more appealing.

The fracture identification, in this case, is based on the rapid breakdown of the electrical pathways caused by the presence of the crack itself. It will be reflected in a dramatic rise in the electrical resistivity of the coating and, hence, in an increase in electrical resistance during the measurement. In this sense, electrical impedance tomography is gaining popularity as a SHM approach. It is based on a mapping of a structure's electrical conductivity based on electrical resistance measurements taken on its surface. As a result, by studying variations in surface resistivity, it will be feasible to detect, localize, and even quantify superficial flaws.

In the framework of polymer coatings, their utility has been broadly established for the detection of superficial cracks [26], where the measurements of electrical resistance between adjacent channels can notably detect artificial harm and sensing skins for spatial pressure mapping. The primary challenge here is electrode positioning and data processing, which usually necessitates the use of complex mathematical algorithms. The results for SHM applications, on the other hand, look promising and add a new capacity to nano-reinforced polymer coatings.

3.5 | Self-Heating of Smart Nano-Composite Coatings

Surface heating is an issue for many applications, and it is typically accomplished through various methods, such as heating elements positioned on the surface or external heating sources, which modify the surface quality of the parts where they are positioned. Heat is necessary in organic coatings for a variety of reasons:

I. Deicing systems: ice accretion on surfaces in humid and cold settings is a regular occurrence that necessitates the use of deicing alternatives, one of which is the heat of the surface to generate a liquid coating.

II. Self-curing coatings: the usage of coatings with curing temperatures above 23°C is generally restricted because of the necessity of external heating sources that develop into a multifaceted production.

However, the glass transition temperatures or hardness are generally higher for higher cross-linked coatings, making the usage of post-curing or higher curing temperatures more appealing.

III. Heat-activated self-healing mechanisms: earlier revealed that the basic sources for self-healing activation are temperature and UV radiation.

On the other hand, the prerequisites for heating limit the adoption of these coatings to structures that can be easily heated. This involves self-healing coatings based on the insertion of conductive nano-particles, such as GNPs and CNTs, into polymer matrices. These coatings are not restricted to the aforementioned applications and have the potential to be implemented in various products requiring heating. Examples include heating seats for commercial vehicles [27], floor heating systems, and heating textiles, where the temperatures required are typically below 100°C [28].

3.5.1 | Fundamentals of self-heating by Joule effect

When carbon nano-particles are mixed into a polymer matrix above the previously described percolation threshold, an electrically conductive substance is formed. The electrical current that travels through the material will generate heat, according to Joule's law, which is commonly referred to as the Joule effect in materials. The first thing to consider is that increased current intensity would result in more heat being generated. Therefore, larger concentrations of carbon nano-particles would be desired for this reason in order to increase the temperature obtained or reduce the voltage required. Although any conventional carbon nano-structure (CNT, CB, even graphite flakes or graphene nano-platelets) may be used for this purpose, high-intensity levels produce the greatest results. In fact, extremely high amounts of CNT have been described in the literature to increase electrical conductivity and, consequently, enable current flow at lower applied voltages. For example, in a study based on ABS as a matrix, CNT was added up to 15 wt. percent to achieve temperatures exceeding 200°C with only 12 V applied [29]. Similarly, research conducted by Chu et al. [30] found comparable temperature and voltage results with 7.5 wt. Percent CNT in PDMS. The use of batteries, which is common in automobiles, has fuelled interest in employing low voltages.

Aside from the heat generated, a significant feature of these percolated electrically conductive networks is the uniform distribution of heat across the covering. For this aspect, two important effects must be considered:

I. Thermal conductivity of polymers is indeed low, thereby making it difficult for heat transfer through the coating.

II. Homogeneous presence of carbon nanoparticles through the polymer matrix is not easily achieved. Separately, CNTs and GNPs have unusually high thermal conductivity.

Despite their identical inherent thermal conductivity, the shape of GNPs makes them more appealing for this application, especially when compared to SWCNT [31]. Despite utilizing the same quantity of both types of nanoparticles, Zakaria et al. [32] found that GNP nano-composites had stronger heat conductivity than MWCNT nano-composites. For instance, whereas greater GNP concentrations are generally required to achieve electrical characteristics comparable to MWCNT, GNP nano-composites displayed a 126.4 percent improvement in thermal conductivity at only 3 wt. % nano-reinforcement, whereas 3 wt. % MWCNT only increased this characteristic by 60.2 percent. Thermal conductivity experimental results are often lower than theoretical values, which have been attributed mostly to waviness, dispersion, alignment, interfacial resistance, and contact resistance [33].

Due to an increase in aspect ratio, proper exfoliation of GNPs results in a substantial increase in heat conductivity. Chu et al. [34] proposed a model for calculating the thermal conductivity of nano-composites based on randomly oriented nanoparticles that considers both the geometrical aspects of the nanoparticles (aspect ratio) and differences in the intrinsic thermal conductivity of the nano-filler in each direction. These properties of GNPs are closely related to their exfoliation and dispersion in the polymer matrix. However, the waviness of the nano-reinforcement may limit the effective aspect ratio of the nano-fillers, suggesting that few-layer GNPs might be an ideal solution compared to individual mono-layers, which tend to roll up easily during the dispersion phase.

These self-heating coatings do not need to have particularly high thermal conductivities, but they should be high enough to assure heat flow throughout the entire facade for the aims earlier indicated. Aggregate formation is prevalent in these materials, which may result in low carbon nanoparticle loadings; certain resin regions are devoid of nano-reinforcement, resulting in non-uniform heating of the samples. In such instances, the sample's thermal conductivity is even more significant because heat is not created uniformly, emphasizing the importance of thermal conduction. When Prolongo et al. [35] compared MWCNT-loaded specimens to GNP-loaded specimens, they discovered that the variations between maximum and minimum temperatures were significantly greater in the MWCNT-based specimens. This effect was also observed with GNPs, as lower GNP contents resulted in areas with lower nano-reinforcement concentration, leading to higher thermal differences between different areas within the same specimen. Conversely, samples containing higher GNP contents (12 wt. percent) demonstrated more uniform heating.

3.5.2 | Deicing system in the form of self-healing

The icing on the structure's surface can substantially impair system performance and potentially cause system damage, necessitating replacement, which raises economic, environmental, and security concerns. Wind turbines and aircraft surfaces are examples where ice accumulation negatively impacts operational conditions by altering the aerodynamic profile, increasing structural weight, and so on [36], [37]. The majority of current techniques are based on two distinct approaches that influence the coatings employed.:

I. Passive techniques that do not require an external source of energy. The fundamental advantage of these methods is that they require less energy to function; however, their efficacy is typically lower. Thus, they are frequently employed in conjunction with active methods to minimize the power required [38]. Hydrophobic coatings are one of the most commonly acknowledged passive methods, which may be accomplished by several approaches, such as the inclusion of nanoparticles or the tuning of surface textures at various length scales, often based on biomimetic techniques [39].

II. Active techniques that necessitate the use of an external energy source. These approaches are often more successful in preventing icing and can be used in conjunction with passive methods. Despite their high power consumption, heating systems are the most dependable of these active approaches, while mechanical ice breakdowns using inflated rubber boots can be discovered in petite planes. When it comes to heating the surface, there are various options (including infrared heating and warm air conduction), but electrical resistance heating appears to be among the most dominant. To prevent the employment of extra membranes or layers, multifunctional coatings with thermo-resistive heating capabilities while protecting the underlying structural material are extremely appealing.

Carbon nanostructures have been researched for this purpose in order to develop an active approach based on Joule effect heating while increasing hydrophobicity by adding nano-reinforcements. Several research initiatives have previously been conducted with this goal in mind, yielding highly promising results in terms of temperature, homogeneity, heart rate, and power consumption. Redondo et al. used GNPs to achieve 35°C temperature increments at 800 V with a heat rate of 13.6°C/min. Even at severe ambient temperatures below 20°C, this temperature increase should be sufficient to cause ice melting on the coating and, as a result, separation of the ice accreted to the surface [40].

When CNTs are employed, their electrical conductivity is generally greater than that of GNPs, raising the value of the intensity at a constant voltage, which is beneficial for generating heat. CNT/PVA films with extremely high CNT concentrations have been created in order to achieve extremely low electrical resistance and, as a result, increased electro-resistive heating. The same method, utilizing high MWCNT loadings (10 wt. percent in poly-1,3,4-oxadiazole), allowed these nano-reinforced films to attain temperatures beyond 100°C with only 40 V applied. Prolonged et al. examined the temperature attained by using CNTs and GNPs as nano-reinforcements and found that with 0.5 wt. Percent CNTs, the temperature reached over 100°C with just 90 V applied, whereas 300 V was required to go beyond 65°C when 8 percent GNP was added [35]. Nonetheless, the scientists reported that when GNPs were utilized as nano-reinforcement, the temperature was more homogeneous.

Lastly, earlier works have engaged in researching the concept of using an electrical network of CNTs to sense temperature and, as a consequence, activate the voltage application when necessary based on weather conditions perceived by the coating. Coatings based on CNT/PDMS were developed and revealed a temperature coefficient with a Negative Influence (NTC). The effect of temperature on electrical response was nonlinear, with sensitivity more than six times higher in the 5 to 10°C range than at room temperature, making them potential candidates for temperature measurement in smart coatings capable of detecting temperature and activating the voltage required accordingly [41].

3.5.3 | Self-curing coatings

Self-heating nano-reinforced polymers have been created based on the same principles as prior applications to allow curing by electrical voltage application. This problem has already been investigated in order to take advantage of the uniform heat created by the material itself, as well as the absence of power losses associated with heat transmission from the heating source to the material itself, as occurs when ovens are employed [42]. Mas et al. demonstrated the efficacy of this procedure by adding MWCNT to an epoxy matrix, and they discovered homogeneous thermal characteristics in the resin cured by Joule heating. They were able to manage the actual curing temperature by attaching thermocouples to a power source and using a PID controller.

The PID used the thermocouple input to regulate the power supply in order to maintain the curing temperature consistent throughout the operation. One of the most significant benefits they discovered was the rapid heating rate caused by heat emitted by the material itself. Jang et al. demonstrated this fact in their study to demonstrate that curing thin films of PDMS reinforced with large loadings of CNT (7 wt. percent) by Joule heating might result in even higher performance than similar oven cured samples. They examined the mechanical characteristics of both materials and discovered that the stiffness of samples treated by Joule effect heating was somewhat greater, owing to quicker and more uniform heating of the entire material volume.

As a result, they conclude that this curing process may allow for faster-curing durations, more homogeneous materials, and greater cross-linked structures [43]. The remaining carbon nanostructures that differ from CNTs have been examined to cure thermoset materials outside of the oven, such as those previously conducted by Mas et al. [42]. GNP usage was advocated to cure epoxy matrices by adding contents more than 8.5 wt. Percent since they discovered this value to be the percolation threshold for the GNP morphology and dispersion technique utilized. They discovered curing degrees comparable to those obtained in oven-cured samples but with significantly quicker heating rates and more homogeneous curing, as in the prior investigations.

Despite this, they identified a preferential orientation of the GNPs, which they attributed to the presence of an electric field during the curing stage because it was not detected in oven-cured samples. This is crucial since they identified improved electrical and mechanical performance in this direction; nevertheless, this anisotropic behavior must be taken into account when designing parts with these materials [44]. Creating remedy by Joule impact of heating has been proven feasible to be electrically conductive networks based on carbon nano-particles inside polymer matrices, which is of particular interest due to faster curing cycles, more uniform curing degree, and ease of application in large structures/surfaces.

3.5.4 Self-healing coatings

The previously described self-heating via thermo-resistive heating might benefit any thermally actuated system. One example is thermo-reversible Diels-Alder reactions, in which the retro-Diels-Alder and Diels-Alders reactions are preferred at different temperatures, allowing the restoration of covalent bonds and, thus, the mending of fractures. Willocq et al. employed this approach to design nano-reinforced polymer matrices using self-healing capabilities via Joule effect heating at low voltages of 25 V, which were sufficient to reach the retro-Diels-Alder reaction temperature in the vicinity of the macroscopic damage (crack) due to local higher heating around the crack due to local higher heating around the crack. This local temperature increase around the fracture was highly dependent on the position of the electrodes in relation to the crack direction [45].

By applying 220 V to the material, Huang et al. [46] developed a percolated network containing GNPs spread in a TPU matrix and achieved 98 percent self-healing efficiency. The use of GNPs, in this case, allowed for the enhancement of other self-healing methods, such as heating via IR radiation absorption, which is also aided by the presence of GNPs in the matrix, allowing for a wide range of options to improve the TPU's thermal self-healing process. This Joule effect-based heating may also be used to initiate self-healing in thermoplastic/thermosetting blends that have self-healing capabilities.

This process is initiated by increasing the temperature above the melting point of the thermoplastic material employed high enough to allow it to flow properly through the fissures formed. Several materials have been researched for this purpose, with Polycaprolactone (PCL) being one of the most commonly studied due to its low melting temperature, which allows a self-healing process to occur at temperatures around 100°C to enable appropriate flow [47]. Zhang et al. [48] demonstrated the effectiveness of adding CNT to EVA/PCL composites for shape memory purposes, allowing the blends to be heated to 100°C, which was sufficient to initiate self-healing processes in blends containing PCL as the healing phase. All of these recent studies show that self-heating can help in the development of self-healing polymer-based materials, with the primary benefits of avoiding the use of external heat sources, employing electrical voltage that can be remotely activated, allowing heat to emerge from the material itself rather than heating other material parts and reducing heat transfer losses.

3.5.5 | Self-cleaning using smart nano-composite coatings

The surface characteristic that maintains cleanliness even in unfavorable environmental conditions is referred to as self-cleaning [49]. The lotus effect inspires it, and it has a low surface energy as well as a micro- and nano-hierarchical surface form that gives superhydrophobicity. The surface is said to be super-hydrophobic if the water contact angle is larger than 150° (WCA > 150°) and the low sliding angle is less than 10° (5A 10°). The use of photocatalytic fillers, which give hydrophilic surfaces (WCA 5–10°) capable of keeping free of organic contaminants and moisture, is another method of generating self-cleaning surfaces. Self-cleaning surfaces may be created using nano-doped polymer coatings and tidy graphitic coatings.

Zinc Oxide (ZnO) and Titanium Oxide (Tio2) are two of the most explored self-cleaning materials due to their photocatalytic and superhydrophobicity breakdown capability of organic pollutants. The sol-gel process is commonly used to generate these coatings. The visible light responsiveness of these nanofillers can be increased by altering them with graphene or its derivatives. Photoexcited carriers can be transferred from TiO_2 to graphene, which narrows the band gap and enhances the material's response to visible light [50]. High interfacial binding energy and hybrid TiO2/graphene nanofillers can exhibit significant electronic overlap, allowing photoexcited carriers to move from TiO2 to graphene and decrease the band gap, increasing visible light photo response. Graphene, on the other hand, enhances the photocatalytic efficiency of ZnO by receiving electrons from ZnO nanoparticles, minimizing the recombination of photo-generated electron holes in the semiconductor. When exposed to visible light, these nanocoating demonstrate superhydrophobicity. Impregnating cotton fibers to produce industrial self-cleaning textiles is an attractive alternative technique for achieving this property. Superhydrophobic coatings are a form of self-cleaning coatings. They involve a hydrophobic polymer matrix, such as fluoropolymers or polysiloxanes, which is enhanced by doping with graphitic nanofillers. These nanofillers provide high electrical conductivity [6]. These coatings exhibit a high contact angle and a low slide angle with water, making them highly effective in repelling water. Additionally, they are very resistant to wear and corrosion . The inclusion of a polymer matrix promotes chemical resistance and improves adhesion to the substrate. When the matrix is not made of a hydrophobic polymer, chemical etching can be used to lower the surface energy of the nanocomposite coating.

This approach enhances the use of a broad spectrum of matrices, such as resin and acrylic, while also looking for additional advantages such as greater adhesion, mechanical qualities, and cheap cost, making it appropriate for a wide range of industrial areas. Along with self-cleaning behavior, various associated features, such as antibacterial, antifouling, anti-soiling, and anticorrosion, are often developed in smart coatings based on carbon nanoparticles. Graphitic nanostructures have demonstrated anti-biofilm and antibacterial properties, although the exact mechanisms involved are not fully understood or well-documented [50]. Other nano-particles of Silver, Gold, Copper, ZnO, TiO2, CuS, and Fe2O3 are anchored to the grapheme nano-sheets for these applications to improve bactericidal efficiency. Currently, efforts are focused on producing membranes and films containing polymer matrices, such as polyethyleneimine and polyamides, for use in disinfection, gas separation, and water desalination.

4 | Conclusion

Polymer films incorporating carbon nanoparticles are becoming increasingly common. They have improved electrical, thermal, and mechanical characteristics that may be exploited to impart new capabilities to them, transforming them into smart materials capable of engaging with their environment and correctly responding to environmental stimuli. The formation of an efficient electrical, thermal, or mechanical network inside the polymer matrix enables these extra activities. For SHM applications, induction resistive heating is used. The Joule effect may be found in self-cleaning and self-healing systems.

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