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Review

Potential of Polymer/Fullerene Nanocomposites for Anticorrosion Applications in the Biomedical Field

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Abstract: Initially, this review presents the fundamentals of corrosion-resistant polymer/fullerene nanocomposites. Then, the potential of polymer/fullerene nanocomposites for corrosion resistance in biomedical applications is presented. In particular, anticorrosion biomedical applications of fullerene-based nanomaterials are proposed for antimicrobial applications, drug delivery, bioimaging, etc. According to the literature, due to the low conductivity/anticorrosion features of pristine thermoplastic polymers, conjugated polymers (polyaniline, polypyrrole, polythiophene, etc.) with high corrosion resistance performance were used. Subsequently, thermoplastic/thermosetting polymers were filled with nanoparticles to enhance their anticorrosion properties relative to those of neat polymers. Accordingly, fullerene-derived nanocomposites were found to be effective for corrosion protection. Polymer/fullerene nanocomposites with a fine dispersion and interactions revealed superior anticorrosion performance. The formation of a percolation network in the polymers/fullerenes facilitated their electron conductivity and, thus, corrosion resistance behavior. Consequently, the anticorrosion polymer/fullerene nanocomposites were applied in the biomedical field. However, this field needs to be further explored to see the full biomedical potential of anticorrosion polymer/fullerene nanocomposites.

Keywords: corrosion; polymer; fullerene; nanocomposite; biomedical

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1. Introduction

Corrosion is a technical issue related to the metallic industries [1]. Several corrosion inhibitors, anticorrosion coatings, and additives have been designed [2–4]. Nanocarbons such as carbon nanotubes, graphene, fullerenes, carbon black, etc. have been applied for corrosion resistance [5]. A fullerene is an important type of nanocarbon that has an exclusive nanostructure and properties [6–8]. Polymers and fullerene nanofillers have been applied to form high-performance nanocomposites [9]. Conjugated polymers have been successfully used for anticorrosion applications. The formation of conjugated polymeric nanomaterials may yield further enhanced corrosion-resistant characteristics. Among the types of nanocarbons, fullerenes have been identified as a unique nanomaterial. Fullerenes may form complex diffusion paths for corrosive molecules to prevent corrosion [10]. The dispersion of fullerenes is considered to have an important effect on the barrier properties of a nanocomposite. Polymer and fullerene interactions, such as π-π-interactions and Van der Waals forces, facilitate nanocomposites' performance. Various methods have been used to form corrosion-resistant nanomaterial coatings, such as solution casting, dipping, spraying, doctor blading, spin coating, printing, etc. [11–13]. In the biomedical sector, the use of anticorrosion fullerene nanomaterials has been found in wide-ranging applications, from bioimplants to drug delivery [14].

Biomedical materials also need corrosion protection to be applied under in vivo and in vitro conditions due to the presence of water/chemical media. In the biomedical sector, recent developments have introduced several solutions for the corrosion protection of biomaterials [15]. In this regard, the introduction of functional nanocarbon-based materials for the encapsulation of corrosion inhibitors and anti-corrosive agents and the modification of biomedical materials via chemical manipulation needs to be implemented. Special attention has been dedicated to functional coatings for the corrosion protection of bioimplants. The mechanism of anticorrosion definitely depends upon the creation of tortuous paths in the materials for the diffusion of corrosive species in biomedical media.

In the area of nanotechnology, there has been tremendous interest in fullerene-based nanomaterials due to their unique properties and flexible dimensional structure [16]. Fullerenes have excellent electrical, thermal, and optical properties, making them promising materials for drug delivery, bioimaging, biosensing, and tissue engineering applications. The surface functionalization or nanocompositing of fullerenes may modify their chemical and physical properties to enhance their drug loading/release capacity, capability of targeting drug delivery toward specific sites, and dispersibility/compatibility in biological systems. Thus, fullerenes have been effectively used in different biomedical applications, including targeted drug delivery, biomedical implants, tissue engineering, wound healing, biosensing, bioimaging, photodynamic therapy, etc.

Conducting polymers and nanocomposites have been used in several technical areas, including that of corrosion protection [17]. Conducting polymers showed better interactions with metal substrates for corrosion prevention compared with those in traditional inorganic coatings [18]. Conducting polymers have superior corrosion-resistance and wear-resistance properties. Compared with inorganic materials, conducting polymers have greater corrosion resistance and non-toxic behavior [19]. Various conjugated polymers have been used for corrosion protection, such as polyaniline, polypyrrole, polythiophene, polyacetylene, etc. [20]. These materials have tailorable conductivity, charge-transfer properties, and environmental stability.

The major principles that can be hypothesized for polymer/fullerene nanocomposites for fine anticorrosion performance in the biomedical field include (i) a surface modification route for fullerene-derived nanocomposites (physical/chemical), (ii) the fine mechanical and structural stability of fullerene-based nanocomposites, (iii) the low cytotoxicity of fullerene-derived nanocomposites, (iv) the tunable surface chemistry of a fullerene nanocomposite layer on bioimplants, and (iv) the antimicrobial properties of fullerene-based nanocomposite. Finally, the perspectives of anticorrosion materials are recognized for biomedical applications.

In this review, the significance of corrosion-resistant polymer/fullerene nanocomposites is discussed, with special reference to biomedical applications. This field needs to be deeply explored to reveal the full significance of biomedical-related anticorrosion polymer/fullerene nanocomposites. Thus, this is an all-inclusive, pioneering, and up-to-date overview of anticorrosion polymer/fullerene nanocomposites, from their design/essential features to their biomedical relevance. To the best of our knowledge, polymer/fullerene nanocomposites for biomedical applications have not yet been extensively reviewed in the literature. Therefore, this review article is doubtlessly a ground-breaking contribution in the field of fullerene nanomaterials intended for advanced biomedical applications. Future developments in the field of polymer/fullerene nanocomposites are not possible for researchers before gaining a knowledge of the literature in this field.

2. Corrosion-Resistant Polymeric Nanocomposites

Polymeric nanocomposites were designed by using polymeric matrices and nanoparticle additives [21]. Polymeric nanocomposites may have superior electrical, mechanical, thermal, barrier, and flame-/corrosion-resistance features [22]. Consequently, polymeric nanocomposites have been applied in technical fields, including in aerospace, automotive, electronics, energy device, construction, and biomedical applications [23–25]. Relative to

the coatings of inorganic materials, conjugated polymers have enhanced anticorrosion features [19]. In this regard, the most commonly used conjugated polymers for anticorrosion comprise polyaniline, polypyrrole, polythiophene, and polythiophene derivatives [26]. Radhakrishnan and co-workers [27] developed poly(vinyl butyral)-, polyaniline-, and nano-TiO_2-based nanocomposite coatings. Electrochemical impedance spectroscopy revealed the better anticorrosion performance of the coatings with >4 wt.% nano-TiO_2. Furthermore, high nanoparticle contents enhanced the anticorrosion characteristics [28]. Moreover, non-conducting polymers were also applied for corrosion resistance. Al_2O_3 nanoparticles improved the scratch/wear resistance of a polymeric nanocomposite [29]. Wang and co-workers [30] examined the tribological and anticorrosion properties of polymer/Al_2O_3 nanocomposites. A nanocomposite with a fine Al_2O_3 nanoparticle dispersion was found to enhance scratch/abrasive resistance. Hence, the success of conjugated polymers, thermoplastics, and thermosetting polymers was analyzed in anticorrosion nanocomposites [31,32]. Conducting polymers may mostly act as cathodes, whereas metals work as anodes via oxidation. This process continues until a metal–polymer interface is developed [33]. Accordingly, various nanofillers have been used as dopants for polymers to improve their conductivity and anticorrosion properties [34].

3. Fullerene

A fullerene is a symmetrical zero-dimensional nanocarbon nanostructure [35]. It resembles a hollow ball structure and consists of sp^2 hybridized carbon. It typically contains a regular arrangement of pentagons and hexagons to form a spherical nanostructure. The fullerene was first discovered in 1985. Among the types of fullerenes, the most common fullerene forms include C_{60}, C_{70}, C_{80}, and others. C_{60} is the most commonly known fullerene and is also referred to as the buckminsterfullerene (Figure 1). Higher fullerene analogs have also been observed with a larger diameter and structure (Figure 2) [36]. Fullerene molecules possess remarkable optical/electrical, mechanical, thermal, and biomedical features [37,38].

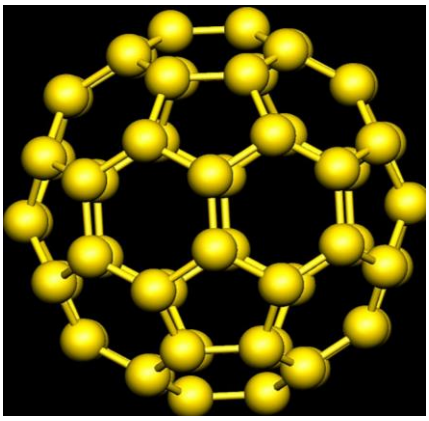

Figure 1. A buckminsterfullerene.

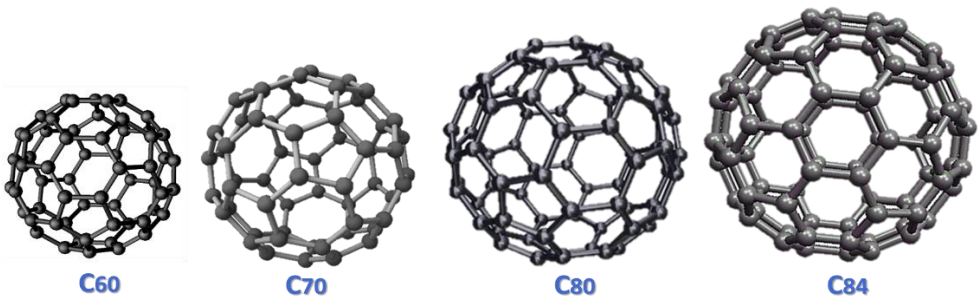

Figure 2. A few higher fullerene analogs.

Fullerenes have been synthesized using various efficient techniques, such as laser irradiation, plasma techniques, microwave synthesis, CO_2 reduction with metallic lithium, and chemical methods [39,40]. Kroto and co-workers [41] synthesized fullerenes through carbon source vaporization by using a laser irradiation method. The subsequent fullerene molecules consisted of clusters of 60 carbon atoms. A plasma technique was used to form good-quality fullerenes [42]. Later, Xie and co-workers [43] proposed the microwave plasma synthesis method to attain fullerenes from chloroform plasma with a low pressure, high temperature, and argon atmosphere. Another attempt by Chen and Lou [44] industrialized C_{60} fullerenes through the reduction of CO_2 by using metallic lithium at a temperature of 700 °C. Moreover, Scott et al. [45] used multi-step chemical conditions to convert a carbon precursor into a C_{60} fullerene. Numerous methods have been used to synthesize fullerene structures. However, all of the techniques have certain relative advantages and disadvantages in terms of the fullerene quality, yield, and processing conditions.

Fullerenes have been used in numerous technological applications, such as solar cells [46], sensors/electronics [47], drug/gene delivery [48], and so on. Fullerenes' solubility has been studied to explore their technical usefulness [49,50]. Polymers have also been used to solubilize fullerene molecules [51,52]. Thus, fullerenes have been identified as unique nanocarbon nanofillers due to their structural properties and the uniqueness of their applications. Like other nanocarbon nanostructures, fullerenes can also be applied in the anticorrosion field. A thin layer of fullerenes was studied for use as anticorrosion coating [53]. So, the inclusion of fullerenes in polymers also enhanced the corrosion-resistance performance of nanocomposites [54].

4. Polymer/Fullerene Nanocomposites for Corrosion Resistance Intended for Biomedical Applications

Corrosion coatings have been used to prevent metal surfaces from surrounding corrosive media [55–57]. The corrosion performance of coatings depends on various factors [58,59]. The inclusion of carbon or metal nanoparticles in polymers further enhances a coating's performance [60–62]. The ensuing high-performance polymer/nanocarbon nanocomposite coatings are inexpensive and lightweight and have a fine processability [63–65]. Fullerene-filled polymer nanocomposite coatings have also been used to increase anticorrosion properties [53,66,67]. The anticorrosion mechanism of a polymer/nanocarbon nanocomposite relies on the formation of a homogenous coating, nanoparticle dispersion, and the generation of diffusion paths for corrosive species [68–70]. Like other nanocarbons, fullerenes have fine anticorrosion features [71]. Fullerenes have the ability to develop electrostatic interactions, π-π interactions, van der Waals interactions, and hydrogen bonds with polymers. These interactions promote charge transfer and the resulting corrosion-resistance properties [72]. Fullerenes have been used to fill various polymers. Among the types of conducting polymers, polyaniline has been significantly used for anticorrosion coatings [73,74]. In the biomedical field, polyaniline has found utility due to its low toxicity, hydrophilicity, high electrical conductivity, biocompatibility, and in vivo stability. However, the uses of polyaniline remain limited due to its low processing capabilities and degradability. In this respect, Zare et al. [75] explored the use of polyaniline-derived nanomaterials in the biomedical sector, such as in biosensors, drug delivery, antimicrobials, and tissue regeneration/engineering. Among the types of thermosets, anticorrosion epoxy coatings have been reported [76–78]. Here, epoxy/nanocarbon nanomaterials have fine hydrophobicity, contact angle, and moisture-resistance properties in addition to their chemical/mechanical/heat stability [79,80]. The anticorrosion properties of epoxy nanocomposites have been widely investigated [81–83]. Thermoplastic polymers have also been composited with C_{60}/C_{70} molecules [84,85]. For corrosion resistance, polyurethane/fullerene nanomaterials were scrutinized [86]. Likewise, poly(methyl methacrylate)/fullerene nanocomposites were also used in corrosion protection [87,88].

In biomedical implants, anticorrosion fullerene nanomaterials have been researched. The most success was found in the case of fullerene-based anticorrosion bioimplants [89,90].

Fullerenes were effectively applied in Mg-based metal implants. In this area, Samadianfard et al. [91] used a sodium dodecyl sulfate surfactant to stabilize fullerene materials prior to nanocomposite formation. The sol-gel method was used to form an anticorrosion coating on the Mg implants. Electrochemical impedance spectroscopy in 3.5 wt.% NaCl was used to detect the corrosion performance of the implants. Scanning electron microscopy and energy-dispersive X-ray spectroscopy, which were conducted at the end of the corrosion tests, revealed that there was the least corrosive damage to the implants' surface, i.e., the surface made of a polymer/fullerene nanomaterial [92]. Later, Jayasathyakawin et al. [93] reviewed the possibility of the use of fullerene nanomaterials for a magnesium (Mg)-based bio-nanocomposite for orthopedic applications. Wang and co-workers [94] studied the cyto-compatibility of nanomaterial-coated bioimplants. The anticorrosion and anti-osteoporosis properties of the bioimplants were studied. In addition, Pourhashem et al. [92] studied the long-term utility and stability of corrosion-resistant fullerene nanostructures in bio-systems. When coated on Mg metal, these anticorrosion fullerene nanomaterials enhanced the biocompatibility and biostability of the bioimplants. Hence, among the biomedical applications of anticorrosion fullerene-based materials, the most success has been seen in the case of the bioimplants.

The antimicrobial properties of carbon nanomaterials have been discussed [95]. Fullerenes have been successfully applied in antibacterial applications [96]. In this respect, Zhao et al. [97] designed conducting and degradable nanomaterials based on polyaniline. Their fine antibacterial activity was observed with respect to bacterial strains such as *E. coli* and *S. aureus* (Figure 3). The in vitro chemical structure, biocompatibility, biodegradation, conductivity, swelling ratio, and rheological properties were studied. Wang et al. [98] reported the antibacterial properties of a corrosion-resistant polyaniline/C_{60} fullerene nanowhisker nanocomposite. The polyaniline-doped C_{60} fullerene nanowhisker revealed homogeneous coating formation over its surface due to the development of a charge-transfer complex (Figure 4). As a result, the antibacterial properties of the nanocomposite were enhanced. Additionally, Ansari et al. [99] studied the antibacterial activity of fullerenol nanomaterials towards Gram-negative/Gram-positive bacterial strains. Subsequently, Liu et al. [100] investigated anticorrosion epoxy/fullerene coatings for anti-bacterial applications. Anticorrosion and tribological studies were performed on the nanocomposite. The properties were found to be enhanced with the loading of fullerenes. Moreover, an important study was reported by Sukhodub et al. [101]. They developed chitosan-, hydroxyapatite-, and fullerene-based anticorrosion materials by using a microwave irradiation method. The antimicrobial activity and anticorrosion performance of chitosan cross-linked with hydroxyapatite and fullerenes were observed against *S. aureus* bacterial strains. More reports are needed in the literature to establish the potential of fullerene-based anticorrosion materials in antibacterial or antimicrobial systems.

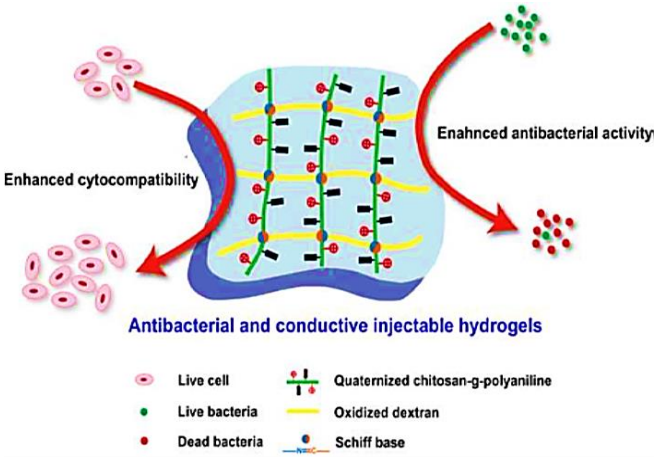

Figure 3. Antibacterial activity of a polyaniline nanomaterial [97]. Reproduced with permission from Elsevier.

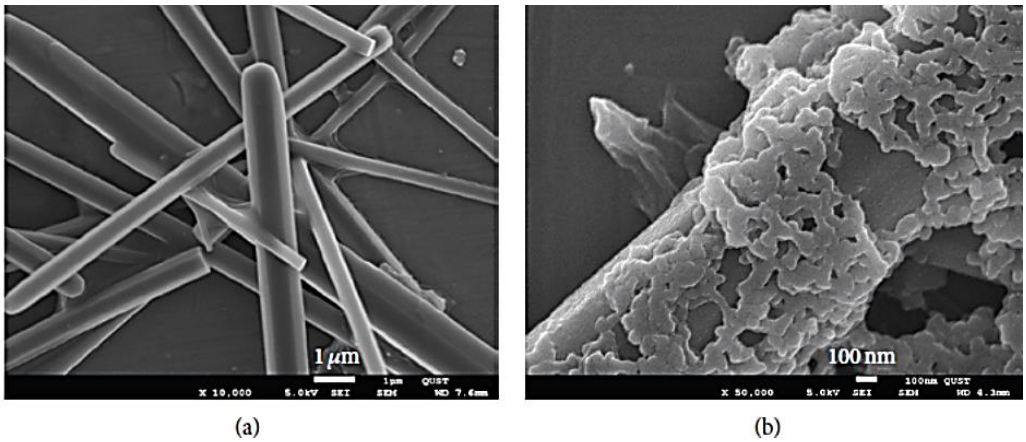

Figure 4. SEM images of (**a**) a fullerene nanowhisker and (**b**) a polyaniline-doped nanowhisker [98]. Reproduced from Hindawi (published with open access).

Fullerene nanoparticles have been used in the drug delivery field [102,103]. Fullerene-derived nanomaterials were found to be capable of delivering chemotherapeutics, nucleic acid, and cardiovascular drugs directly to cancerous cells. An important study by Hasiao et al. [104] explored the use of polyaniline/poly(lactic-co-glycolic acid) nanomaterials for drug delivery (Figure 5). Simulation studies on the electrical conductivity and cytotoxicity were performed for the nanomaterials. Subsequently, Thong et al. [105] synthesized polyaniline- and fullerene-based nanocomposites for drug delivery. The nanocomposites also had anticorrosion properties. Polyaniline was used in the emeraldine and leucoemeraldine forms. More polymer/fullerene nanocomposite designs have been observed for drug delivery applications [106]. C_{60}-based molecules were found to be useful in drugs for resisting bacteria, fungi, and pathogens [107]. Later, Biswas et al. [108] designed a silver-nanoparticle- and fullerene-based nanomaterial for effective anti-cancerous drug delivery. Anticorrosion properties are desirable for preventing dose-dependent toxic effects in vitro. The use of silver-nanoparticle-decorated C_{60} endo-fullerenes with antimicrobial and anticorrosion properties could bring revolutionary changes in the field of drug treatment regimes. Again, more efforts are required in the literature to see the in vivo and in vitro toxic effects and anticorrosion performance of fullerene-based materials in drug delivery systems.

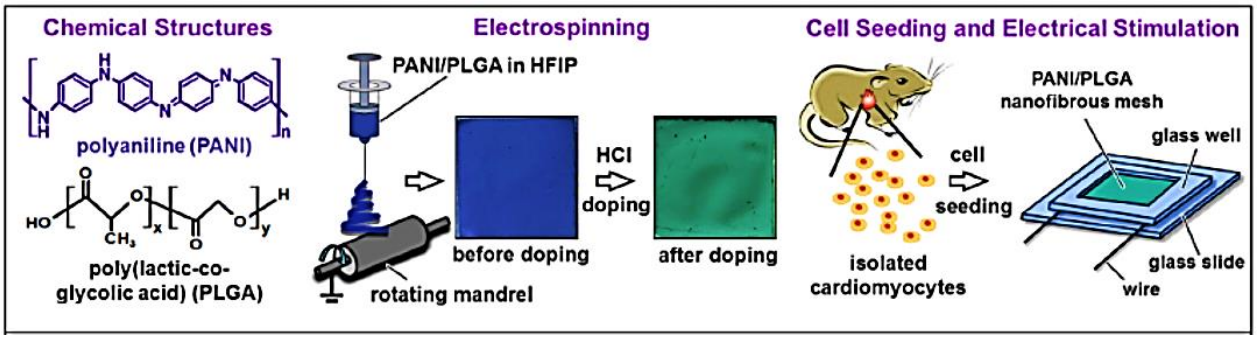

Figure 5. Graphic for the fabrication of aligned conductive polyaniline (PANI)- and poly(lactic-co-glycolic acid) (PLGA)-based nanomaterials for drug delivery [104]. Reproduced with permission from Elsevier.

Fullerenes and fullerene-derived anticorrosion nanostructures have been applied in bioimaging [109]. In this regard, endohedral fullerene derivatives were proven to be effective for bioimaging. Figure 6 demonstrates an endohedral fullerene as a bioimaging agent. Consequently, the final biomedical properties of anticorrosion polymer/fullerene nanocomposites depend on the polymer type, fullerene functionalization, nanofiller content,

dispersion, and matrix–nanofiller interaction [110–112]. Although the bioimaging field has directly used fullerene-based anticorrosion materials, there have been limited attempts to chart this research field.

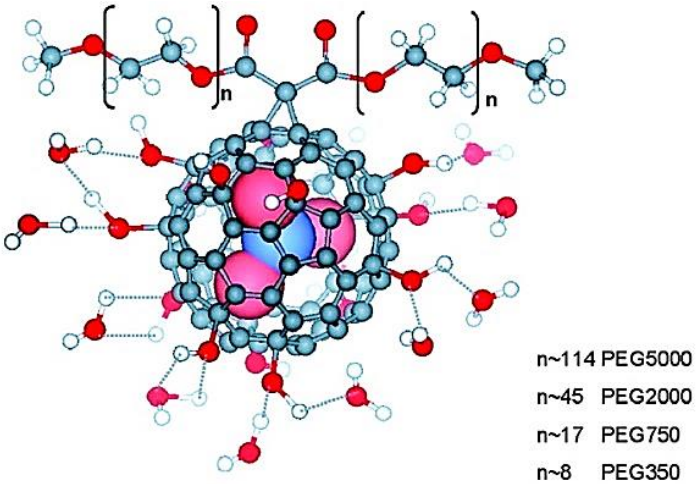

Figure 6. Depiction of the chemical structure of a $Gd_3N@C_{80}[DiPEG(OH)_x]$ endohedral fullerene particle. $Gd_3N@C_{80}[DiPEG(OH)_x]$ = Trimetallic nitride/fullerene/dipoly(ethylene glycol) hydroxide [109]. Reproduced with permission from ACS.

5. Future Challenges and Summary

In this review, the biomedical applications of fullerene-based nanocomposites were thoroughly investigated (Figure 7). In this regard, the use of anticorrosion polymers, polymeric nanocomposites, and polymer/fullerene nanocomposites was considered. Fullerene-reinforced polymeric nanocomposites have been employed for the development of high-performance corrosion-resistant materials. The resulting polymer/fullerene nanocomposites displayed superior corrosion-resistance performance due to their high surface area, light weight, high electrical conductivity, and corrosion-resistance performance [113,114]. Conjugated polymer/fullerene nanocomposites revealed high corrosion resistance, but non-conducting polymer/fullerene systems were also used for anticorrosion purposes [115]. Anticorrosion polymer/fullerene nanomaterials were used for technical applications [116]. These coatings had the ability to block corrosive species, including moisture, chemicals, gases, etc., by creating barrier effects. Finally, polymer/fullerene nanomaterials with a fine nanofiller dispersion may enhance anticorrosion properties [117].

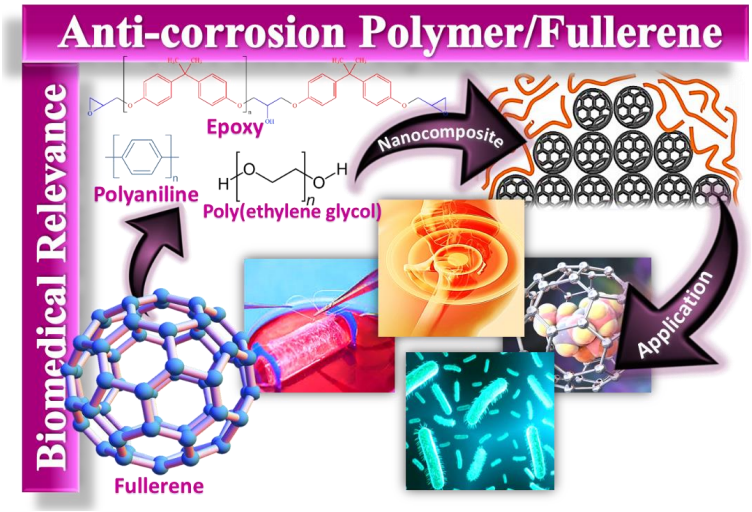

Figure 7. Significance of corrosion-resistant polymers/fullerenes.

Since the discovery of the fullerene, this family of symmetric nanostructures of nanocarbon nanomaterials has opened new research eras for technical fields. The incredible properties obtained through the combination of polymers and fullerenes have gained more attention in the field of advanced nanomaterials. Hence, fullerenes and the derived nanomaterials have become subjects of curiosity for the biomedical engineering community. Due to their exceptional physical and chemical properties, fullerenes have been proven to be a favorable option for applications in biological chemistry. In this regard, various challenges have been identified for the use of fullerenes and the derived materials in biomedical applications.

First of all, it has been found to be essential to fabricate the modified or functional form of a fullerene. One obstacle or challenge in using fullerenes in biomedical applications is their insolubility or low solubility in water. π-conjugated unsaturated double bonds have been found to be effective in developing functional fullerene molecules. In addition to fullerenes, fullerene polymers have been developed to enhance the functional structures of such nanocarbons. Incidentally, fullerene-containing polymers (polyfullerenes) have been developed, and they have better solubility properties. In particular, polyfullerenes may have linear, cross-linked dendrimers and end-capped fullerene polymers.

Another challenge in using fullerenes is their toxicity and biocompatibility. In this respect, as nanocarbons, fullerenes have the ability to accumulate in intracellular spaces, so their toxic effects have been studied. The toxicity of fullerenes may reveal different results in biomedical applications depending on their deteriorated physiochemical properties and ROS-related behavior. Here, processing methods and chemical modifications may alter the general properties of pristine fullerene molecules and the derived nanomaterials. Various studies have been carried out to explore the toxic effects of fullerene nanomaterials on human mammalian cells. Although fullerenes are not deadly nanomaterials, acute or sub-acute toxicity has been observed in living systems.

Now, the use of anticorrosion fullerenes in biomedical applications also faces challenges. Regarding fullerenes used in biosensors, anticorrosion fullerene materials have been found to be important for detecting microorganisms, enzymes, biomolecules, antibodies, etc. However, the corrosion of sensing materials may cause several problems in biological systems. As efficient mediators for sensors to conjugate with biomolecules include hydrophilic and functional groups for electron transfer, the chances of corrosion increase. Therefore, anticorrosion polymer fullerenes must be applied in biosensors. Again, the issues of fullerene materials' solubility and functionalization have been considered.

In magnetic resonance imaging (MRI) for diagnosis, anticorrosion fullerene materials have gained importance. In this regard, endohedral fullerenes with entrapped metal atoms in cage-like structures (which are not released in in vivo environments) act as contrast agents in magnetic resonance imaging and X-ray imaging. Correspondingly, the anticorrosion properties of fullerene nanomaterials have been found to be important due to their easy water uptake properties. For MRI imaging, the most successful anticorrosion material developed so far is the poly(ethylene glycol)-/fullerene-based photosensitizer, which has dual applications in diagnosis and treatment.

The biomedical applications of fullerenes have gained research attention due to their antioxidant activity and radical scavenging properties. Corrosion may cause toxic effects that lead to free radical production, abnormal reactions, tissue abnormalities, and abnormal cellular metabolism.

In drug delivery, fullerene-based nanomaterials have been used in delivery methods, such as oral administration or injection. Here, the corrosion of in vivo nanomaterials has been found to be challenging. Moreover, the possibility of corrosion may limit cell activity and the overall cell metabolism. In this respect, corrosion-resistant nanomaterials for targeted drug delivery may control the drug release and biocompatibility properties without causing any corrosion-related toxicity.

Although there are still major issues to overcome and the use of fullerenes and corrosion-resistant fullerenes, polymers/fullerenes in biomedical engineering will certainly have a bright future.

In a nutshell, this review was developed while keeping in mind the importance of anticorrosion polymer/fullerene nanomaterials in biomedical systems. The significance of anticorrosion polymer/fullerene coatings was specifically investigated for the emerging biomedical applications. However, research in these areas is still in an infant stage, and future efforts are required to develop advanced polymer-/fullerene-nanocomposite-based biomedical materials. At this point, conjugated, thermoplastic, and thermosetting polymers have been used to form anticorrosion nanomaterials with fullerenes in the biomedical field. In this regard, functional fullerene molecules need to be used to form more biocompatible and efficient anticorrosion nanostructures. Consequently, polymer/fullerene nanocomposites have been successfully designed for bio-implants, tissue engineering, drug delivery, anti-microbials, and other biomedical applications. However, all of these areas need to be thoroughly explored by using new design innovations, novel processing approaches, and new analysis techniques.

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