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Review

Fullerene in water remediation nanocomposite membranes—Cutting edge advancements

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Abstract: Among carbon nanoparticles, fullerene has been observed as a unique zero-dimensional hollow molecule. Fullerene has a high surface area and exceptional structural and physical features (optical, electronic, heat, mechanical, and others). Advancements in fullerene have been observed in the form of nanocomposites. Application of fullerene nanocomposites has been found in the membrane sector. This cutting-edge review article basically describes the potential of fullerene nanocomposite membranes for water remediation. Adding fullerene nanoparticles has been found to amend the microstructure and physical features of the nanocomposite membranes in addition to membrane porosity, selectivity, permeation, water flux, desalination, and other significant properties for water remediation. Variations in the designs of fullerene nanocomposites have resulted in greater separations between salts, desired metals, toxic metal ions, microorganisms, etc. Future investigations on ground-breaking fullerene-based membrane materials may overcome several design and performance challenges for advanced applications.

Keywords: fullerene; nanocomposite; membranes; water remediation; permeation

1. Introduction

Benefits of membrane skills have been observed for technical utilization due to low cost, efficient working, low energy consumption, and scaled-up processing [1]. In traditional membrane technologies, pressure-driven membrane assemblies have been used for filtration [2]. Developments in this field have led to the implication of polymer-based membranes and materials. Consequently, numerous polymers have been used as filtration membranes to enhance their robustness, selectivity, permeability, and desalination performance at low pressure [3,4]. Polymeric membranes have been fabricated using a range of techniques, such as simple solution casting, phase inversion, sol-gel procedures, and efficient electrospinning techniques [5–7]. The efficiency of polymeric membranes has been found to be reliable on the surface area, microstructure, porosity, crystallinity, hydrophilicity, etc., enhancing flux, fouling resistance, and desalination performances [8–10]. In this regard, applying nanocomposite membranes instead of pristine polymeric membranes has been found advantageous [11]. Consequently, carbon nanostructures like graphene, carbon nanotubes, nanofibers, etc. have been employed to develop nanocomposite membranes [12,13]. Most competent fullerene nanoparticles have been applied to polymeric membranes [14]. Hence, water remediation has been investigated using polymer- and fullerene-derived nanocomposite membranes [15–17].

This state-of-the-art article presents fullerene-filled nanocomposite membranes for water purification. Inclusion of fullerene in filtration membranes caused significant effects on the separation performances owing to the high surface area, pore size, porosity, surface roughness, and other surface properties [18]. Fullerene-based membranes revealed fine potential to overcome the performance challenges of the filtration of unwanted pollutants [19].

2. Fullerene

Fullerene is a hollow, symmetrical carbon nano-allotrope with sp^2 hybridization [20,21]. Owing to structural features, π conjugation has been observed in the fullerene molecule [22]. This cage-shaped nanostructure has a size of about 1 nm. Its discovery dates back to 1985 [23]. Fullerene molecules have been found as C_{20}, C_{24}, C_{60}, C_{70}, C_{120}, etc., depending upon the number of carbon atoms in the hollow ball-like ring structure (**Figure 1**) [24]. Fullerene C_{60} is the most frequently adopted form, known as buckminsterfullerene. This marvelous molecule has been studied for its optical, electronic, mechanical, thermal, and biomedical properties [25]. A number of techniques have been used to form the fullerene molecules, like the plasma method, chemical vapor deposition, arc discharge, and many others [26,27]. Advancements in fullerene research have been observed in the form of nanocomposite structures [28,29]. For nanocomposite formation, the solubility of fullerene molecules has been considered [30]. Various solvents like water, poly(vinylpyrrolidone), and organic solvents have been used for fullerene molecules [31]. Consequently, better-processed fullerene nanomaterials have been applied for photovoltaics, optoelectronics, sensors, and biomedical applications [32–35]. Furthermore, high-performance fullerene-based nanocomposite membranes have been designed. The membrane performance was dependent upon the type of fullerene molecules, dispersions, and interactions with the matrix materials used [36].

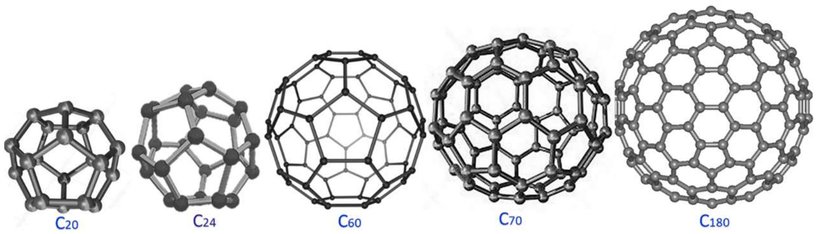

Figure 1. Some fullerene molecules.

3. Nanocomposite membranes

Various technological sectors have focused on the application of membranes [37,38]. In this regard, membranes have been effectively applied for the removal of environmental contaminants from water [39]. Most importantly, polymer-based nanocomposite membranes have been designed with numerous potential benefits for water separation [40]. Accordingly, the pollutants from ground, domestic, sea, and industrial water have been removed using the advanced membranes [41]. The membrane filtration efficiency definitely relies on the permeability and selectivity features [42]. Moreover, nanocomposite membranes have been explored for improved physicochemical properties [43,44]. Important membrane features studied in this regard include porosity, hydrophilicity, selectivity, fouling, mechanical, and heat stability [45]. A range of different filtration nanocomposite membranes have been prepared, such as microfiltration, nanofiltration, ultrafiltration, reverse osmosis, mixed matrix, and so on [46,47]. The membrane properties also depend on the nanofiller type, quantity, and dispersion features of the polymeric systems [48]. For nanocomposite membrane formation, various nanocarbon nanoparticles have been used, including graphene, carbon nanotubes, nanodiamonds, etc. [49,50]. Similarly, wide-ranging polymers have been adopted to form efficient membranes [51]. For example, reports on polysulfone- and graphene-based nanocomposite membranes have been observed [52,53]. The polysulfone/graphene nanocomposite membranes were fabricated using the phase inversion technique [54]. These membranes have been investigated for crystallinity, morphology, and matrix-nanofiller interactions, enhancing their physical properties and water remediation performance [55]. Similarly, countless polymer/nanocarbon nanomaterials have been reported for membrane applications.

4. Fullerene in nanocomposite membranes for water remediation

Fullerene-filled nanocomposite membranes have been prepared and examined for membrane properties like desalination, toxic ion removal, metal ion removal or recovery, and microorganism separation from water [56]. Various toxic metals like lead, mercury, arsenic, etc. have been removed using the efficient fullerene-filled membranes [57–59]. The separation performance of these membranes relies on the porosity and surface defects of these membranes [60,61]. Perera and colleagues [62] reported on fullerene-based reverse osmosis membranes. The membranes revealed a high water flux of 26.1 L/m^2h and salt rejection properties. The nanocomposite membranes were effectively used to separate the lithium ions from seawater [63].

Polyamide is a commodity thermoplastic polymer with amide bonds in the main chain [64,65]. Polyamide has been effectively adopted for membrane application [66,67]. Plisko and co-researchers [68] designed the polyamide and hydroxy functional fullerene-derived nanocomposite membranes for water remediation. Adding 5 wt.% nanofiller aided the antifouling properties. In addition, the removal of organic matter has been observed for the nanocomposite membranes. Dmitrenko et al. [69] used polyamide polyphenylene isophthalamide and filled it with fullerene nanoparticles along with other carbon fillers. The mixed matrix pervaporation membranes have been fabricated through the solid-phase synthesis method. **Figure 2** displays a simple route for the formation of polyphenylene isophthalamide/C_{60} pervaporation membranes. The inclusion of nanofiller increased the transport properties of the nanocomposite membranes. The membranes were tested for the transport properties of an azeotropic methanol-toluene mixture. Adding fullerene nanoparticles has considerably improved the permeation flux of the membranes [70]. Here, permeation flux was observed in the range of 0.084–0.214 $kg/(m^2h)$ with 5 wt.% fullerene contents. In addition, a selectivity of 96 wt.% was observed. The porosity, permeability, and selectivity of the pervaporation membranes were dependent on the fullerene contents and interactions with the polymers [71,72].

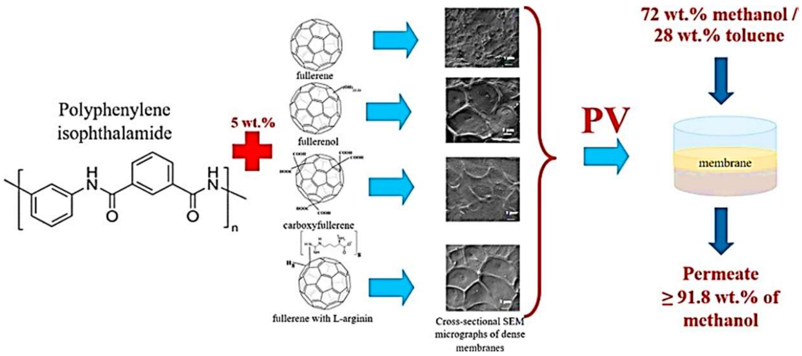

Figure 2. Graphical representation of development of novel polyphenylene isophthalamide pervaporation (PV) membranes modified with various types of C_{60} derivatives [72]. Reproduced with permission from MDPI.

Liu et al. [73] reported on epoxy-derived nanocomposite membranes filled with fullerene C_{60} and graphene oxide. The resulting membranes have been studied for their ion permeation and desalination properties. **Figure 3** shows a transmission electron microscopy micrograph of fullerene and graphene oxide-based nanomaterials. The interlayer spacing between the fullerene-grafted graphene nanosheets was found to be around 100 nm due to the insertion of 0.7–1 nm fullerene nanoparticles. Due to interlayer spacing, a low permeation rate was observed. **Figure 4** expresses the fabrication and water desalination setup for the formation of water permeation membranes of epoxy and fullerene-grafted graphene nanoparticles. Including fullerene molecules led to a high water flux of up to 10.85 L/m^{2}hbar. Better desalination and water permeation have been observed. **Figure 5** displays the variations in ion concentrations on permeation vs. time for the fullerene-based membranes. The stability features of the nanocomposite membranes were found to affect the desalination performance [74]. **Table 1** exhibits examples of some fullerene-filled nanocomposites-based filtration membranes.

Table 1. Specifications of few polymeric membranes with fullerene nanofiller for water purification.

Nanoparticles	Fullerene nanoparticle size (nm)	Membrane pore size	Filtration (L/m^{2}h.bar)/LMH.bar	Ref
C_{60}	14–59	34 to 55 nm	-	[68]
Functional C_{60}	~1	0.86 to 0.59 nm	26.1 LMH	[62]
Polyhydroxylated C_{60}	-	0.64 nm	6.7 LMH.bar	[63]
C_{60}	0.14	-	-	[75]
C_{60}	-	Large pore size	-	[76]
C_{60}	-	17 nm	-	[77]
C_{60}	9–15	5 wt.% nanoparticles small pores	0.084–0.214 kg/(m^{2}h)	[69]
C_{60}	0.375	-	-	[78]

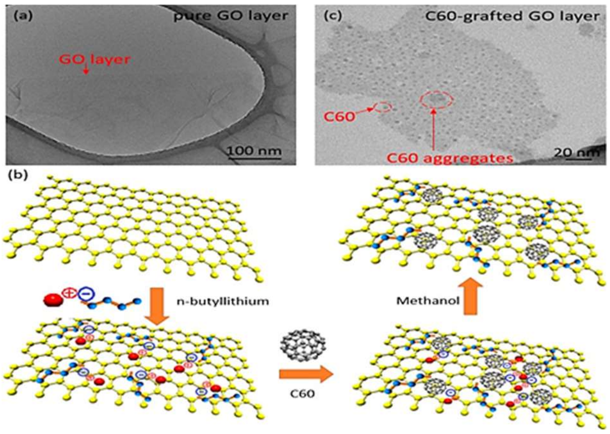

Figure 3. (a) Transmission electron microscopy (TEM) image of pure GO layer (very thin layer with a little folding edge represents GO layer, at scale bar of 100 nm); **(b)** Schematic illustration of grafting C_{60} on GO layer through lithiation reaction; and **(c)** TEM image of C_{60} grafted GO layer (smooth layer with irregular shape represents GO layer and dark dots represent C_{60} nanoparticles, at scale bar 20 nm). The GO layer is around 150 nm, whereas the C_{60} nanoparticles are 1–2 nm) [73]. GO = graphene oxide; C_{60} = fullerene. Reproduced with permission from ACS.

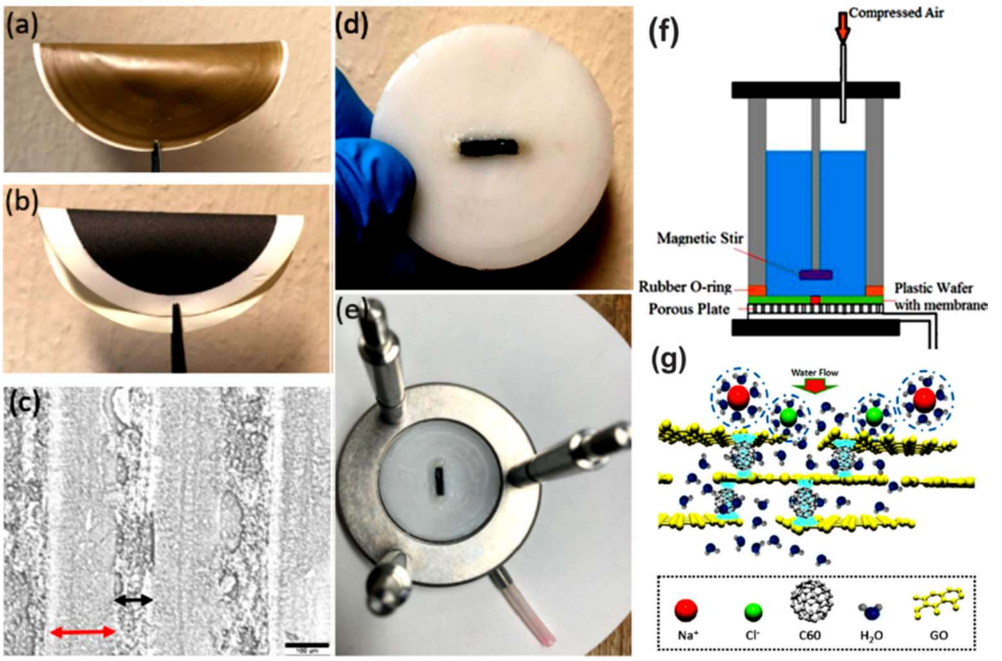

Figure 4. Fabrication process and water desalination setup using C_{60} grafted graphene oxide membranes. The photograph shows: **(a)** graphene oxide membrane without C_{60}; **(b)** C_{60} grafted graphene oxide membrane; **(c)** optical micrograph of cross-sectional area with scale bar 100 µm. The micrograph shows 148 µm thick graphene oxide laminates embedded in 81 µm thick epoxy; **(d)** graphene oxide-C_{60} membrane encapsulated with epoxy in plastic disk of 47 mm; **(e)** graphene oxide-C_{60} membrane inside water desalination setup; **(f)** and **(g)** are schematic setup of flat membrane made of graphene oxide and C_{60} hybrid for water desalination [73]. GO = graphene oxide; C_{60} = fullerene; Reproduced with permission from ACS.

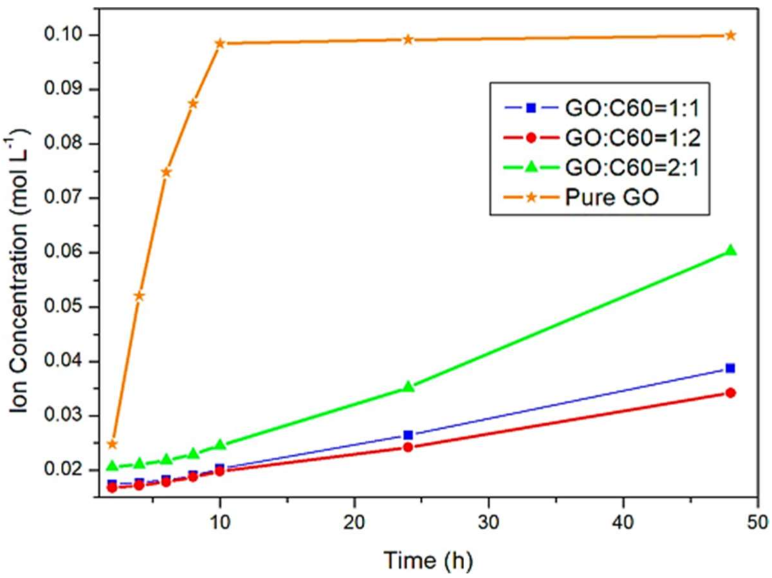

Figure 5. Ion concentration on the permeation side through GO/C_{60} membrane over time period (the red, blue, and green lines indicate the feed ratios of GO:C60 = 1:2, 1:1, and 2:1, respectively) [73]. GO = graphene oxide; C_{60} = fullerene. Reproduced with permission from ACS.

Polysulfone is a marketable thermoplastic polymer commonly used [79]. Polysulfone has several advantageous features, like chemical, mechanical, and thermal robustness. Polysulfone has been used to form membranes, coatings, and other practical nanostructures for methodological fields [80]. Penkova and colleagues [81] reported on polysulfone and fullerene-derived mixed-matrix membranes. Adding 5 wt.% fullerene C_{60} enhanced the membrane transport features, especially pervaporation of the ethyl acetate-water mixture [82]. Including fullerene nanofiller also elevated the membrane surface area and hydrophilicity. The solution-diffusion processes were used to promote pervaporation through the membrane [83]. Consequently, mass transfer and permeability were found to increase through the membranes.

Nafion is another important matrix for membrane formation [84,85]. Nafion-based commercial membranes have been widely adopted for environmental, energy, and energy/electronics applications [86,87]. Here, fullerene-filled nafion membranes have been produced [88]. The antimicrobial properties of the nanocomposite membranes were considered. Tasaki and colleagues [89] formed the nafion/fullerene nanocomposite membrane using the solution casting method. The solvent technique was efficient in forming compatible fullerene-filled membranes [15]. The membranes were studied using molecular dynamic simulations, and fine fullerene nanoparticle dispersion was deliberated. Layon et. al. [90] developed fullerene nanocomposites using poly(vinyl pyrrolidone) as well as different solvent media. The resulting membranes were used for wastewater remediation. **Figure 6** shows that the sonication technique better dispersed the fullerene nanoparticles in the medium relative to aqueous dispersion and in tetrahydrofuran. Fullerene nanoparticles had a size of 30–100 nm. In poly(vinyl pyrrolidone), aggregated fullerene nanoparticles have been observed [91]. The effects of minimal inhibitory concentrations on aggregate surface area can be seen in **Figure 7**. There was no linear relationship between the minimal inhibitory concentrations and aggregate surface area. However, enhanced surface area increased membrane performance due to better interactions.

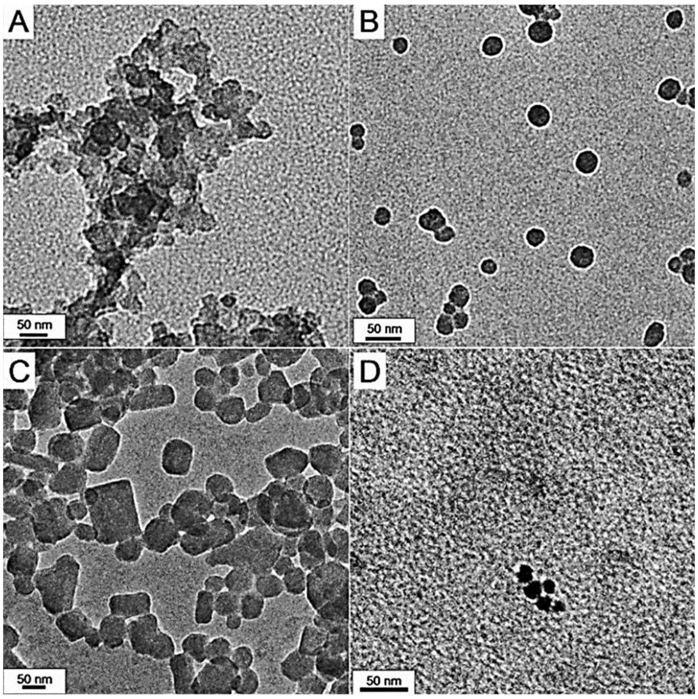

Figure 6. Transmission electron microscopy micrographs of **(A)** aq/nC_{60}; **(B)** son/nC_{60}; **(C)** THF/nC_{60}; and **(D)** PVP/nC_{60} [90]. Reproduced with permission from ACS.

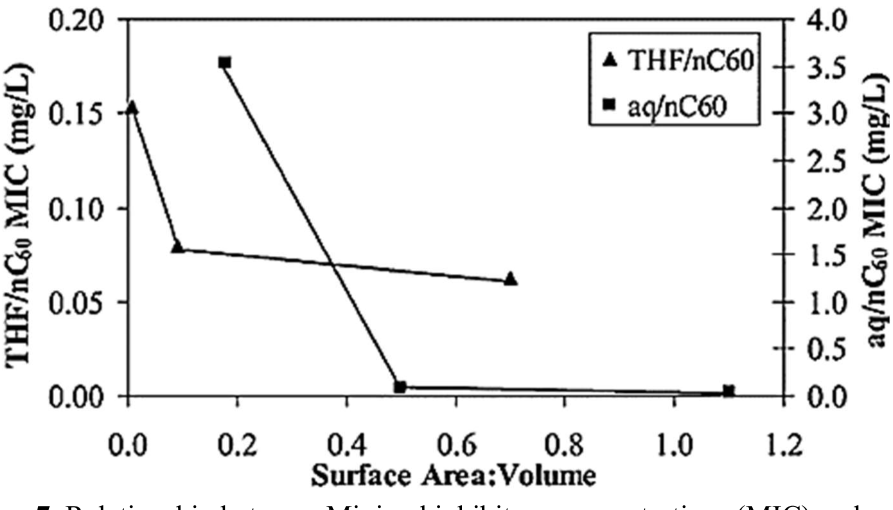

Figure 7. Relationship between Minimal inhibitory concentrations (MIC) and aggregate surface area. There is no linear relationship between the mean MIC and the surface area to volume ratio calculated, indicating that the difference in surface area alone does not account for the difference in MIC between the small and large aggregates [90]. Reproduced with permission from ACS.

5. Prospects and conclusions

Fullerene nanostructures have brought about revolutions in a range of methodological industries, including organic photovoltaics, energy, biomedical purposes, biopharmaceuticals, etc. [92–94]. Fullerene nanocomposite membranes have been widely used in filtration systems. Other water decontamination strategies have also been considered, such as sedimentation, distillation, biological processes, flocculation, chlorination, ultraviolet light, etc. [95]. Various combinations and types of polymer/fullerene membranes have been developed (**Figure 8**). In fullerene-based membranes, remarkable morphology, mechanical, and barrier features have significantly contributed towards water remediation [96]. Fullerene molecules have contributed to the matrix-nanofiller interactions, enhancing the compatibility of these nanostructures. The main challenging aspect has been recognized as nanoparticle dispersion in polymeric membranes [97].

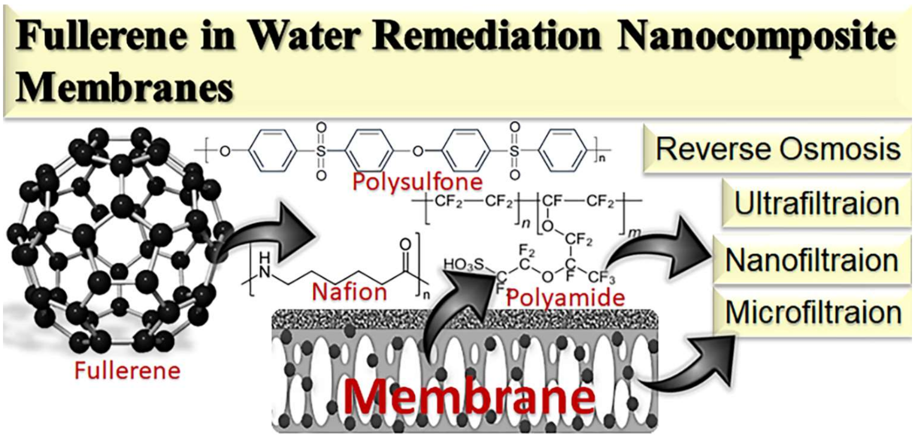

Figure 8. Design of fullerene-based membranes.

Better fullerene dispersion throughout the membrane ultimately defines the controlled pore size or structure, morphology, surface roughness, and wettability of efficient membranes. In this regard, separation mechanisms need to be explored to further improve the fullerene membrane-based filtration processes. Theoretical studies on fullerene nanocomposite membranes may also help to resolve the performance challenges. In the future, variations in membrane designs may also bring about revolutions in this field.

This cutting-edge review presents an analysis of applying fullerene nanocomposite membranes for water purification purposes. Polymer-based nanocomposite membranes with fullerene nanoparticles have been found to transform waste water remediation. Efforts on fullerene nanocomposite membranes have led to improved surface properties, permeability, selectivity, separation, antifouling, and other features. These membranes have a low price and lasting stability for large-scale filtration. Further research may lead to a number of enhanced membrane parameters to overcome these drawbacks.

Conflict of interest: The author declares no conflict of interest.

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