Application of Metal/Metal Oxide Doped Electrospun Nanofiber Membranes in Sustainable Catalysis

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Abstract

The unsustainability of the production of catalysts due to limited resources and higher energy demands makes it critical to explore and reengineer new catalytic materials for future applications. Woefully, the unrelenting demand for the metals/metal oxides increases both the financial and environmental cost, particularly in mining and synthesis, rendering consumption unsustainable in its current form. In this context, electrospinning offers a new template for designing sustainable ways of minimizing the higher loading of catalysts and recyclability. In this context, metals/metal oxide doped electrospun membranes have grasped a great scientific interest as sustainable catalysts due to their enhanced catalytic activity and synergistic structure-property relationship of the doped material and the matrix. More specifically, the selectivity arising from the electronic properties and quantum mechanical interactions at the nanoscale of metal/metal oxide nanoparticles coupled with interactions at the electrospun membrane interfaces lead to such enhanced properties. This review article summarizes the applications of metals/metal doped electrospun membranes in different aspects of catalysis, such as thermocatalysis, photocatalysis, organocatalysis and electrocatalysis, with a particular focus on their sustainability.

Keywords: Sustainable catalysis, metal/metal oxide doped catalysts, electrospinning, organocatalysis, photocatalysis

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1. Introduction

Driven by the need to pursue sustainable development goals recommended by the United Nations based on green chemistry principles, over the past several decades, the interest in academic and industrial research and innovations have shifted towards developing more environmentally-friendly practices and sustainable solutions(Cespi et al., 2020). Catalysts have always played a crucial role in attaining many critical demands, such as avoiding large amounts of chemicals, increased selectivity to reduce waste generation and decreased energy demand propelling the research towards greener routes. Catalysts have become indispensable in most refining and petrochemical processes, energy production, as well as in the synthesis of specialized chemicals and, more recently, in addressing environmental challenges, particularly for preventing pollutant contamination by enhancing reaction selectivity to prevent waste by-products (Védrine, 2019).

Mostly, the precious metals and metal oxides in powder forms are the powerhouses of the catalysts. When using metal-based catalysts, metals seeping into the environment is a major problem in many applications, particularly in large-scale industrial manufacturing, making recovery of these valuable catalysts difficult and requiring costly post-treatment measures(Shao et al., 2012, Munaweera et al., 2014b). When metal catalysts are attached to a substrate, the quantity of leaching is reduced to a negligible level, resulting in less product contamination and pollution. Furthermore, recovering the costly metal catalysts on a substrate can be as easy as filtering the reaction mixture in most situations or employing a magnet for catalyst separation in others(Ghasemi et al., 2015). For this catalyst loading, one-dimensional nanofibers have been widely employed in the construction of substrates, and it has been demonstrated that recovering and recycling these nanofibrous catalysts has minor to no influence on their catalytic activity(Lu et al., 2019a, Munaweera et al., 2014a).

Electrospinning has recently garnered great scientific interest in catalysis research due to its great potential in producing continuous one-dimensional nanofibers with an interconnected pore structure(Rezaee et al., 2017). Fibers exhibit different physical and chemical properties at the nanoscale that are not observable at the macro scale. Nanofibers, for example, have a substantially larger specific surface area, resulting in enhanced activity in catalytic applications due to the augmented accessibility of the reactive surfaces to the reactants in the chemical process(Lu et al., 2015), enhancing the efficiency of electron-hole pair separation (Wang et al., 2018), and the interconnected porous fibrous structure facilitates efficient charge and mass transfer during catalytic activity. Electrospinning also allows fine-tuning the surface chemistry of the fiber by manipulating the functionality of the polymer and hence, gives access to improve the selectivity of the membranes towards the targeted reactants providing intrinsic activity between the reactants(de Oliveira Santos et al., 2017). Furthermore, the blending of the metal/metal oxide nano-catalysts during polymer processing assists in uniform dispersion of the catalysts throughout the membrane preventing the agglomeration.

Although there have been myriad approaches employed to fabricate catalyst incorporated electrospun membranes they can be classified into two categories as; blending through the electrospinning process or post-electrospinning deposition (Nasir et al., 2021). In electrospinning encapsulation, the catalyst or the catalyst precursor is blended into the polymer solution prior to electrospinning. This will produce an electrospun nanofiber mat with these catalysts encapsulated within the fiber matrix. The catalytic nanofiber mat will then be used with or without further treatments. If the catalytic precursor is encapsulated within the polymer matrix, further processing, for example, chemical activation, calcination or reduction, of the mat is made to incur the active catalyst(Pei and Leung, 2015). In the post-electrospinning deposition method, first, the nanofiber membrane is electrospun without the active catalyst, and then the catalyst or the catalyst precursors are deposited or attached using a number of techniques. The catalyst nanoparticles are in-situ synthesised by reducing the deposited catalysts precursors, which result in much higher catalyst-support interactions that surpass the interaction between the preformed catalysts deposited by simple physical interactions(Moreno et al., 2015). Atomic layer deposition(Zhao et al., 2017), magnetron sputtering(Li et al., 2021), electrospraying(Huang et al., 2017), and vacuum filtration(Zhang et al., 2021a) are a few methods for post-deposition of metal/metal oxide catalysts on electrospun membranes allowing the formation of hierarchical nanostructures which will enhance the catalytic activity of the membrane(Kayaci et al., 2014). Combinational electrospinning and post-treatment methods have not been limited only to polymer nanofibers, but inorganic metal oxide nanofiber composites have also been developed for catalytic applications(Jung et al., 2012). These metal

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oxides are mixed with a polymer and electrospun into a fibrous mat, which is then exposed to heat treatments or calcination to preserve the metal oxide as a fiber after the degradation of the polymer.

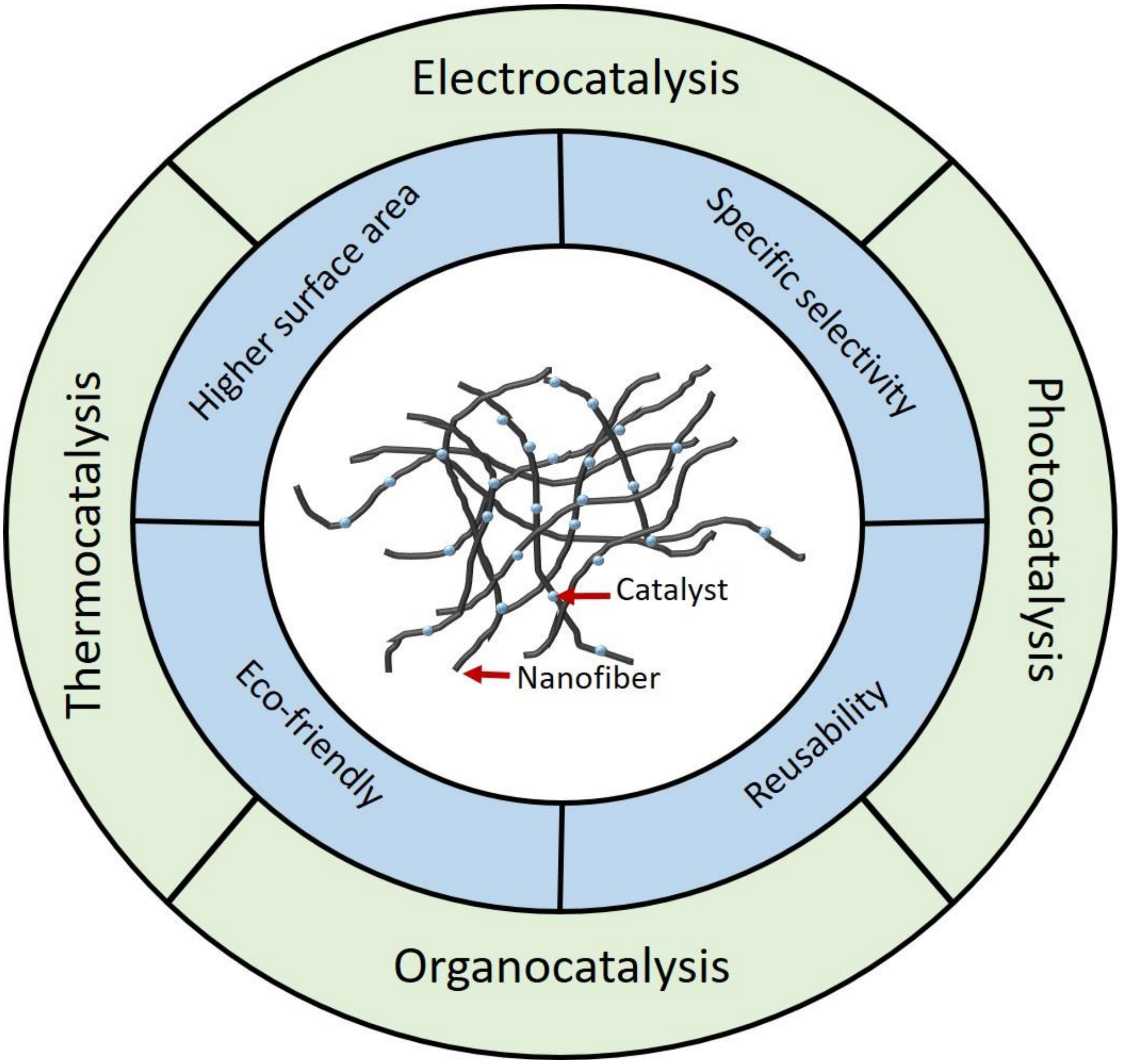

Figure 1. Graphical illustration of the applications and benefits of sustainable electrospun catalysts.

Electrospun membrane supported catalysts have shown a considerable increase in catalytic activity, stability, selectivity, and reusability and have been used for photocatalytic, organocatalytic, thermocatalytic or electrocatalytic applications. As such, electrospinning membrane supported catalysts hold great potential for use in many applications such as chemical synthesis, environmental remediation, and energy production. This review will summarize the recent developments of electrospun nanofiber/ metal/metal oxide catalyst composites and their use in various applications in industrial and environmental applications. (Fig 1) The progress of the current research will be followed by a brief section of future directions of these distinguished metal/metal oxide doped electrospun membranes.

2. Sustainable Catalysis

Despite the fact that metals/metal oxides are commonly used in catalysis, there are issues with their continuous and mass use in catalytic processes. For one thing, the fact that metals/metal oxides are scarce implies that they are in limited supply, are extremely expensive, and are subject to supply variations, all of which lead to growing concerns about their long-term availability. Antimony, the platinum-group metals (osmium, rhodium, palladium, iridium, platinum, and ruthenium), mercury, and tungsten are estimated to be at the highest risk. The constant demand for low-abundance metals/metal oxides not only generates economic concerns but also has severe environmental ramifications. Lower-grade metal ores are mined and refined, resulting in increased fossil fuel use and CO_2 emissions. (Das and Mohanty, 2019). Due to the limited economic viability of these techniques, it is estimated that less than 1% of precious metals are recycled for reuse, despite their great demand and scarcity. As a result, current research concentrates on the ability to make a transition of the current industrial processes away from precious metals, which are finite resources, and toward more sustainable alternatives. In this regard, scientists conducted numerous studies to reduce the amount of metals/metal oxides utilized in catalytic reactions. As a result, the phrase "sustainable catalysis" was coined in response to growing worries about the depletion of precious metals, emphasizing the importance of combining economic and social progress with environmental preservation. In this regard, the current approach for the development of more sustainable catalysis is the minimizing of use of metals and increases the catalytic efficiency by the manipulation of matter at the nanoscale(Quesne et al., 2019).

Because of the numerous beneficial uses in catalysis, porous membranes with smart and functional surfaces have piqued scientific interest. Metal/metal oxide nanoparticle immobilization, which is particularly effective for catalysis, can also be supported by porous membranes. The substrate should ideally have a large specific surface area, high catalyst loadings, and porosity for reactant access in order to obtain high catalytic efficiency(Bac et al., 2020).

The porous membrane structure not only allows for a larger surface area for chemical reactions but it also aids in the dispersion of catalytic substances, resulting in products that do not need to be processed further. Moreover, high catalytic efficiency can be achieved due to the manipulation of metal/metal oxides on the nanoscale. Further, metal/metal oxide doped electrospun nanofibers exhibit a remarkable catalytic efficiency because of the large available active surfaces. It is possible to recover the catalyst particles, which is generally difficult in the chemical process sector. Among different membranes, metal/metal oxide doped electrospun nanofibers have received distinguished attention in the various organic synthesis, environmental remediation and energy production applications in recent years, and it has been briefly discussed in the next sections.

2.1. Application of metal/metal oxide doped electrospun nanofiber membranes in organocatalysis

Metal/metal oxide doped electrospun nanofibers play an integral role in organic catalysis since those nanofibers are mostly used as supports for the immobilization of the catalytic-active species such as metal or metal oxide nanoparticles. Two approaches can be applied to achieve efficient organocatalysis through metal/metal oxide doped electrospun nanofibers. First, the precursor of the catalytic species can be found in the doping material of electrospinning. In this situation, spinning results in the development of composite nanofibers, which are then electrospun to produce the necessary metal/metal oxide doped nanofibers. Catalytic species are stabilized on nanofibers in the second technique. Organic catalysis based on metal/metal oxide doped electrospun nanofibers-based catalysts can be utilized to promote a wide range

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of chemical transformations as well as photo and electrocatalysts. The examples of recent advances in metal/metal oxide doped electrospun nanofibers used in organic synthesis are mentioned in Table 1.

Table 1: Recent advances in metal/metal oxide doped electrospun nanofibers used in organic catalysis.

Electrospun nanofiber composite	Doped metal/ metal oxide	Catalyzed reaction	Reference
TiO_2 and ZrO_2 nanofibers	Platinum (Pt), Palladium (Pd), Rhodium (Rh)	Suzuki coupling reactions	(Formo et al., 2009)
Polyvinylpyrrolidone (PVP) crosslinked electrospun membranes	Palladium (Pd) and Cu_2O	Heck, Suzuki (PVP–Pd) and click chemistry (PVP–Cu_2O) reactions	(Savva et al., 2014)
Polyacrylonitrile (PAN) nanofibers	Titanium dioxide (TiO_2)	Degradation of methanol in gas-phase	(Modesti et al., 2014)
Electrospun Al_2O_3 membranes	Copper (Cu)	Bisphenol A degradation	(Wang et al., 2017c)
Poly(vinylpyrrolidone) (PVP) electrospun membranes	Copper (Cu)/Selenium (Se) nanoparticles and reduced Graphene Oxide (rGO)	Reduction of 4-nitrophenol (4-NP) to 4-aminophenol (4-AP)	(Yang et al., 2015)
Free-standing Ce–W–$TiO_{2-\delta}$ nanofibrous membranes	Titanium dioxide (TiO_2)	Removal of NOx in a selective catalytic reduction (SCR) process	(Dankeaw et al., 2019)
TiO_2 nanofibers	reduced Graphene Oxide (rGO)	Filtration-enhanced highly efficient photocatalytic degradation with the membrane	(Gao et al., 2020)
Polyacrylonitrile (PAN) nanofibers	Zeolitic imidazolate frameworks (ZIF-67)	Activation of peroxymonosulfate (PMS) as a catalyst to degrade yellow-17 (AY) color pigment oxidative catalytic degradation of other pollutants for the recovery of wastewater, proving the decomposition of 68.3% of tetracycline (TC), 100% of bisphenol A (BPA), and 98.3% of	(Wang et al., 2017a)

		rhodamine B (RhB) by ZIF-67/PAN	
Metal–organic frameworks (MOF) BIT-58 (BIT stands for Beijing Institute of Technology) nanofibers	Lanthanide (La)	Knoevenagel condensation	(Chen et al., 2018)
Polypropylene (PP) nanofibers	Aluminium oxide (Al_2O_3), Zinc oxide (ZnO), or Titanium dioxide (TiO_2)	Fast catalytic degradation of chemical warfare agents (CWAs) simulant.	(Lee et al., 2017)
poly-ϵ-caprolactone (PCL) fibers	Platinum (Pt)	Hydrogenation of cyclohexene	(Leus et al., 2018)
MIL-53(Al)-NH_2 nanofibers	Aluminium (Al)	Knoevenagel condensation	(Liang et al., 2018)
SiO_2 nanofibrous	Manganese dioxide (MnO_2)	Cooperate with hydrogen peroxide to form a Fenton-like reagent for the degradation of methylene blue (MB)	(Wang et al., 2017b)
Electrospun poly(vinyl alcohol) (PVA) /poly(acrylic acid) (PAA) /carboxyl-functionalized graphene oxide membranes	Silver (Ag) nanoparticles	Photocatalytic degradation of MB	(Liu et al., 2018)
β-phase polyvinylidene difluoride (PVDF) nanofibrous membrane	Platinum (Pt) np	Electrocatalysis as Oxygen Reduction Reaction (ORR) catalyst	(Zhang et al., 2014)
TiO_2–carbon composite nanofibrous membranes	Vanadium (V)	Complete oxidation of low-concentration acetone	(Chen et al., 2015)
electrospun ceramic nanofibers, such as TiO_2, SnO_2, and ZrO_2,	Platinum (Pt)	Hydrogenation of azo bonds in methyl red	(Ahmaruzzaman and Mishra, 2021)
Carbon nanofibers	Palladium (Pd)	catalytic degradation of an organic dye (MO) in the presence of sodium borohydride	(Najem et al., 2020)
Electrospun polyvinyl alcohol (PVA)/polyethylenimine (PEI) nanofibers	Aurum (Au)/Silver (Ag)	Reduction of 4-nitrophenol to 4-aminophenol	(Hu et al., 2018)
Polyethyleneimine/Polycaprolactone Composite Fibers	Palladium (Pd)	Reductions of 4-nitrophenol (4-NP)	(Wang et al., 2019)

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		and 2-nitroaniline (2-NA)	
Cellulose acetate nanofibers	Silicon (Si)/Tungsten (W)	Decomposition of tetracycline (TC) and methyl orange (MO)	(Li et al., 2017)
Polyacrylonitrile (PAN) and nylon based electrospun membranes	Palladium (Pd)/ Aurum (Au)	Selective Oxidation of 5-Hydroxymethylfurfural	(Bonincontro et al., 2020)

2.2 Application of metal/metal oxide doped electrospun nanofiber membranes environmental remediation

With the continuous industrial growth and urbanization, environmental pollution has become a critical issue and has raised the attention of researchers to find solutions. The pharmaceutical, textile, food processing, paper making and agricultural industries are the major culprits that release large scale environmental pollutants and toxins that cause adverse effects not only to humans but also to the ecosystems (Kumari et al., 2020). However, there are various approaches toenvironmental remediation such as adsorption, ozonation, electrochemical filtration, biodegradation etc. Catalytic degradation of the contaminants has become a topic of interest among researchers due to the demand for renewable energies to achieve green chemistry routes(Zakria et al., 2021, Kumarage et al., 2022). Researchers have utilized the photo-catalytic, electrocatalytic and thermocatalytic ability of the metals and metal oxides for the degradation of the pollutants into harmless products.

In water treatment, the catalysts are usually dispersed in contaminated water, resulting in expensive post purification steps to separate the catalysts after use. To overcome this issue incorporation of these catalysts onto a substrate has become the state-of-the-art. In photocatalytic degradation, TiO_2 is the most exploited metal oxide owing to its photostability, low cost, eco-friendliness, non-toxicity and acceptable bandwidth (Buso et al., 2008). However, TiO_2 lacks adsorption selectivity towards essential organic pollutants in the presence of other organic elements. This results in increased scavenging of reactive oxygen species (ROS) by the unwanted organic elements(Seneviratne et al., 2021). Coupling of TiO_2 with a suitable substrate aids in the adsorption of the prioritized pollutants near the photocatalytic sites and allows the efficient use of short-lived ROS(Singh et al., 2013). For instance, Lee et al. have reported TiO_2 embedded PVP and PVDF electrospun bi polymer system for the photocatalytic degradation of organic contaminants recognized as endocrine disruptors, BPA and 17α-ethynylestradiol (EE2)(Lee et al., 2018). It has shown a 96% removal for both BPA and EE2 in water within 4 hrs and 1.5 hrs, respectively. They have detailed that the TiO_2 embedded electrospun mat has shown several potential advantages over the conventional TiO_2 slurry system. The use of polymer mat doesn’t need any complex separation process, which also prevents the unwanted release of TiO_2 into the water and offers easy recovery of the TiO_2. In addition, the hydrophobic polymer surfaces adsorb and concentrate organic matter closer to the active site, which facilitates and speeds up the photocatalysis of the contaminants. In another study, it was also reported that due to a tight link between the TiO_2 layer and the PVDF substrate, PVDF-TiO_2 core-shell electrospun nanofiber mats displayed steady performance even after five cycles of methyl orange deterioration(Yin et al., 2021). They have post-treated the electrospun PVDF membranes dipped in Ti precursor with microwave-assisted hydrothermal treatment to produce PVDF-TiO_2 core-shell electrospun nanofiber with catalytic photo-oxidation activity. Xu, et al. has reported photocatalytic regeneration of polyacrylonitrile/ titanium dioxide/ polyaniline hybrid electrospun membrane after adsorption of Congo red dye and Cr^{6+} heavy metal(Xu et al., 2020). During regeneration, the adsorbed Congo red will be photo degraded while Cr^{6+} will be partially reduced to Cr^{3+} and desorbed through the photocatalytic process. However, TiO_2, although it has been extensively applied for its photocatalytic activity, it has its own demerits. The band gap of 3.2 eV is comparatively large, limits the electron transfer and is photoactive only under UV light which is only a 4% of the total sunlight limiting its applications(In et al., 2007). In order to overcome this limitation, the use of co-catalysts along with TiO_2 has been proven to be efficient. In this regard, Zhang et al. have deposited TiO_2 decorated molybdenum disulfide (MoS_2) nanosheets on electrospun polyacrylonitrile membrane by vacuum filtration method(Zhang et al., 2021b). The membrane had shown degradation greater than 80% for five different organic dyes. A PAN/ β-cyclodextrin (β-CD) membrane modified with TiO_2 and GO evaluated for photocatalytic degradation of both cationic (MB) and anionic (MO) dyes had greater than 90 % dye degradation capability for both the dyes. The composite has shown 80% degradation capability even after 3 cycles and also has shown good antimicrobial properties against *E. coli* and *S. aureus*.(Zhang et al., 2021b). Here the addition of TiO_2 and GO has decreased the fiber diameter of the electrospun membrane. They have uniformly dispersed and attached the TiO_2 onto the GO sheets by ultrasonic cavitation rendering the nanofibers with the best mechanical properties and barrier properties. The adverse effects caused by the antibiotics released into the environment cannot be underestimated. Due to their complex structures, although they are found in low concentrations, antibiotics have high water stability and genotoxicity and have the capability of developing resistant pathogens(Cuerda-Correa et al., 2020). Zhao et al. developed a nanofiber membrane using modified triaxial electrospinning loaded with β-FeOOH and TiO_2 for the photodegradation of doxycycline(Zhao et al., 2021). The degradation ability was improved by the strong electron-hole conductivity and the matching band gap energies. The membrane has shown a photodegradation efficiency of 90.14% within 5 h. TiO_2 modified electrospun polymer membranes have not only been used for water treatment but for air purification. Electrospun carbon nanotubes(CNT) /TiO_2 nanofibers has been evaluated for its photocatalytic degradation of gaseous benzene of 100 ppm and has exhibited a 52% degradation efficiency under visible light irradiation within 90 mins(Wongaree et al., 2016). The membrane was first fabricated by electrospinning PVP loaded CNT and TiO_2 in required proportions and then removing PVP by calcination at 450°C. The membrane has also exhibited a degradation efficiency of 58% for MB dye in water. By improving specific surface area, lowering band gap energy, and minimizing electron recombination, CNT had a synergistic impact on the increase of photocatalytic activity for CNT/TiO_2 nanofibers. Another air filter for the simultaneous filtration of particulate matter and photocatalytic degradation of toluene has been synthesized by Su et al. by blending TiO_2 with PAN nanofibers via simultaneous electrospinning of the polymer and electrospraying of TiO_2 suspension(Su et al., 2017). This methodology has widely dispersed TiO_2 to form hierarchical nanostructures on the PAN surface and has provided the composite with strong photocatalytic activity along with high filter efficiency. The protrusions caused by the nanoparticles on the polymer membranes increase the effective surface area while increasing the surface roughness of the membrane.

Another widely used catalytic metal oxide used for environmental remediation is ZnO. Due to the porous nature of nanofibers, earlier studies showed that ZnO nanoparticles and nanofiber membrane composites outperformed ZnO nanoparticles in catalytic degradation of organic dye molecules and will also enhance the membranes' antifouling properties(Samanta et al., 2015). For the first time, Pascariu and the group have reported a hybrid electrospun PVDF membrane loaded with lanthanide (Ln=Sm, La, Er) doped ZnO (ZnO:La) nanoparticles for adsorption and photocatalytic degradation of dye organic pollutants under visible light irradiation(Pascariu et al., 2021). The highest colour removal efficiency of the hybrid membrane with 37% ZnO: La was 96.33 % for MB dye degradation and 93.36 % for RhB degradation. Furthermore, even after 10 repeated cycles, the materials could be recovered and reused, with a photocatalytic activity of up to 98 percent. The incorporation of ZnO: La has also increased the mechanical stability of the PVDF membrane compared with the pristine PVDF. In most research the ZnO

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has been evaluated for its photocatalytic performance, but it has been found that the electrocatalytic degradation is much higher than photocatalytic degradation. The electrocatalytic efficiency of ZnO nanofiber membranes is influenced by the size, dispersibility, and stability of ZnO particles(Bera et al., 2016). A PVDF electrospun membrane with ZnO nanoparticles deposited via magnetron sputtering had shown a more than 95% removal of several organic dye contaminants when an electric field of 30 V was applied(Li et al., 2021). Ji et al. have developed an electrospun PAN air filter with parallel side by side nanofibers (SBS NFs) with ZnO and Ag for organic contaminant removal with antibacterial properties. They have electrospun separate ZnO NPs/PAN and Ag NPs/PAN solutions with parallel nozzles into one membrane. The filter has shown 97% degradation efficiency for MB within 140 mins and antibacterial activity against gram-positive and gram-negative bacteria. They have also compared a single nanofiber (SNFs) membrane, which was synthesized by electrospinning a blend of ZnO and Ag Nps in PAN and has revealed that the SBS Nf has a higher filtration efficiency compared with the SNFs membrane.

Platinum (Pt) is another efficient thermocatalytic metal to oxidize air pollutants at room temperature(Zhang et al., 2012). Formaldehyde (HCHO), which is a common indoor air pollutant, causes irritations in the eye/skin and also causes cancers when overexposed, even at low concentrations(Collins et al., 2001). Pt nanoparticles supported on a TiO_2 porous hierarchical nanofiber (Pt/TF) mat synthesized by electrospinning followed by precipitation have shown a much higher catalytic activity of HCHO oxidation into CO_2 and water compared to powder like Pt/P25 catalyst. The pristine mat, however, had the disadvantage of brittleness which has been overcome by supporting the Pt-Ni nanoparticles on polymethyl methacrylate electrospun fibers with a coating of polypdopamine (He et al., 2009).

The fibrous catalyst has shown a much higher removal of HCHO than the powder catalyst and also has exhibited a significantly lower dosage for similar catalytic activity. Other groups have used the thermocatalytic activity of cheaper metal oxide such as Co_3O_4 and MnO_2 supported with electrospun PAN instead of expensive noble metals for the optimum removal of HCHO at temperatures of 98°C and 60°C, respectively (Wu et al., 2016).

2.3 Application of metal/metal oxide doped electrospun nanofiber membranes in energy production

In the face of rising energy scarcity and environmental concerns, photocatalytic water splitting to hydrogen (H_2) and oxygen (O_2) has been viewed as an exciting and promising technique to create sustainable, clean energy at a low cost with a realistic efficiency. Hydrogen as an energy resource plays a vital part in future energy solutions for delivering pollution-free and carbon-free power for buildings, transportation, and industry. As given below, the photocatalytic hydrogen evolution reaction (PHER) (equation 1) and the photocatalytic oxygen evolution reaction (POER) (equation 2) are the two halves of the photocatalytic hydrogen evolution process from water.

$$2e^- + 2H^+ \rightarrow H_2 \qquad \text{equation 1}$$

$$2H_2O + 2H^+ \rightarrow 4H^+ + O_2 \qquad \text{equation 2}$$

Hydrogen evolution photocatalytic materials must have strong responsiveness to visible light across a broad range of wavelengths, efficient charge separation and substantial surface area in order to achieve high hydrogen generation efficiency under solar light irradiation(Liu et al., 2019). To fulfil all these characteristic features, TiO_2 has been blended with co-catalysts to increase its light responsiveness which is limited only to UV light when applied alone, and high surface area TiO_2 nanofibers have been developed in order to increase the charge separation efficiency. For instance, Wang et al. (Wang et al., 2018) developed a carbonated doped TiO_2 nanotubes using olive oil-assisted electrospinning method followed by calcination for PHER. Doping of carbonate has efficiently narrowed the bandgap of TiO_2, extending the photoactive wavelength range of TiO_2 from UV to visible light. Another commonly used bimetallic oxide is $BiFeO_3$ (BFO), for its merits of appropriate bandgap energy for solar light harvesting, strong oxidizing ability, efficient charge separation due to intrinsic spontaneous polarization and built-in electrical field, low toxicity and abundance. With these distinctive properties, Mo doped BFO (Mo-BFO) composite has been developed with a tubular structure for photocatalytic water splitting via electrospinning and subsequent thermal treatment(Zhang et al., 2020). Here Mo has been substituted in place of Fe of BFO. This substitution has boosted the photo-electrical efficiency and enhanced the interfacial charge transfer efficiency between the catalyst and the electrolyte. They have further promoted the POER by applying an amorphous carbon coat on Mo-BFO porous nanotubes. Sn^{4+} doped BFO has also been synthesized via electrospinning and subsequent annealing treatment(Ren et al., 2018). The nanofiber composite with 1% Sn doping, under visible light illumination, has shown an improved catalytic water splitting and photostability. A novel architecture of discrete heterojunction nanofibers has been used for enhancing the photocatalytic activity and charge-separation efficiency by Tao and group. They have developed Bi_2WO_6 nanosheets grown on electrospun BFO nanofibers using a solvothermal technique (Tao et al., 2020). The hybrid has shown 31.9 times greater POER compared to BFO fibers alone and is credited to the charge separation with axial transport of the photogenerated charges and provision of a substantial amount of reactive sites owing to the high surface area conducive to the nanoscale heterojunction. Furthermore, the ferromagnetic properties of BFO allow the magnetic recyclability of the composite. Co_3O_4 nanofibers derived by oxidative heat treatment of electrospun PAN/ Cobalt acetate have been employed for thermocatalytic H_2 evolution from NH_3BH_3(Figen and Filiz, 2019). The optimum H_2 evolution rate achieved by the composite was 2.54 l H_2 min^{-1} g cat^{-1} and the catalyst has exhibited good repeatability.

Direct conversion of solar energy to electric energy using dye-sensitized solar cells (DSSC) is a highly efficient green energy producers that have piqued the interest of academics and industries due to its low cost and ease of implementation. A DSSC consists of four main components as; a transparent photoanode, a monolayer of dye sensitizer, an electrolyte with a redox couple and a counter electrode(Gong et al., 2012). The photoanode is a glass sheet processed with a conductive oxide layer and deposited with a mesoporous oxide layer for active electronic conduction. A monolayer of dye molecules is then covalently attached to the anode for improved light harvesting. Next to the dye sensitizer, a layer of electrolytes in an organic layer is kept for the regeneration of dye molecules. And the counter electrode, which is a glass-coated with a catalyst, is implemented for the collection of electrons.

The mesoporous oxide layer of the photoanode of the DSSC is responsible for electronic conduction activation, being a media for the electron transfer from the dye molecules to the glass sheet and being a good substrate to hold the dye molecules in place. Hence, the photoanode should have the merits of a porous morphology with a high surface area, an appropriate bandgap energy and high electron mobility(Andualem and Demiss, 2018). Considering the foregoing, several semiconductors (such as TiO_2, ZnO, and SnO_2) have been extensively researched and employed as the oxide layers on the photoanode of DSSCs. To improve electron mobility and transit speed, one-dimensional electrospun TiO_2 nanofibers and their blends with nanocrystalline TiO_2 have been explored(Song et al., 2005). The pristine TiO_2 1D nanofibers lack efficient dye molecule adsorption on their surface. In account of its increased light adsorption via enhanced Mie scattering without reducing dye adsorption, these blended composites can attain an improved DSSC performance. Joshi et al. (Joshi et al., 2010) has achieved a 44% higher efficient performance of DSSC by introducing a photoanode with a composite containing 15% nanofibers and 85% nanoparticles of TiO_2. The demerit of inefficient attachment of the dye molecules onto these 1D TiO_2 nanofibers has been overcome even by surface modifying the fibers. For instance, Sun et al.(Sun et al., 2020), has developed Ag decorated TiO_2 electrospun nanofibers for efficiency enhanced DSSC. The

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power conversion efficiency of the Ag decorated DSSC has increased by 18% when compared to pristine TiO_2 nanofiber photoanode.

The counter electrode's primary functions are collecting electrons, regenerating electrolytes, and providing optimal electrocatalytic performance for the redox pair. Carbon nanofibers (CNF) have been suggested as a promising candidate for the electrocatalytic support to be employed as the counter electrode of the DSSC due to their strong corrosion-resistance, large specific area, and fibrous and porous structure that enable high charge mobility. Platinum (Pt) electrodes being the most extensively utilized electrodes due to their outstanding electrocatalytic ability and conductivity, Pt modified with Ni_2P has been incorporated into CNF using two-step electrospinning and redox method to be used as the counter electrode(Zhao et al., 2017). The composite has exhibited excellent electrocatalytic activity and an improved electrical conductivity with a power conversion efficiency of 9.11%. However, the exorbitant cost and low electrochemical stability of Pt, fuelled the demand for a more cost-effective, corrosion-resistant, Pt-free counter electrode material(Lu et al., 2019b),(Jiang et al., 2019). In this regard, Ma et al.(Ma et al., 2019), has developed a Pt free CNF with Fe-Co nanoparticles by electrospinning followed by post-treatments as a counter electrode for DSSC, and the photovoltaic efficiency of the electrode was 98.5% than that of the Pt electrode. They have attributed this enhancement to the good conductivity of the Fe-Co/CNF electrode, the increased contact area between the electrolyte and the electrode due to the 3D mesoporous network and the higher number of active sites due to the high surface area of fiber structures.

3. Conclusion and Prospects

The challenge of switching to a greener, more sustainable chemistry will inevitably necessitate a radical transformation of many aspects of chemistry, particularly all aspects of catalysis and applications. From the standpoint of green chemistry, this entails reengineering catalytic materials that: make the most efficient use of catalytic materials, such as metal/metal doped nanoparticles, which have higher catalytic activity than their equivalents and are simple to synthesize without extensive procedures. All these objectives will necessitate the development of innovative and efficient catalysts that can be generated in a sustainable manner without producing unacceptable quantities of metal/metal nanoparticles. The urgent need for society to transition to a greener, more sustainable future presents a very exciting opportunity for catalysis in a variety of fields, including biomedical applications, organo synthesis, and environmental aspects. The directed design of new catalysts, which requires a detailed theoretical understanding of the activity of metal/metal doped electrospun membranes, will drive many of the necessary changes in resource management and increased energy efficiency. Electrospun membranes containing minimal concentrations of metal/metal oxide nanoparticles have been discovered to be an excellent tool for developing long-term catalysis in a variety of applications. The remarkable thing about these membranes is that their catalytic properties are mostly due to their high surface area-to-volume ratio of solid-supported metal particles. Furthermore, as has been proven for many catalytic systems, the activity and selectivity of isolated nanoscaled catalytically active phases is enhanced in these metal/metal doped electrospun membranes. Furthermore, since this equipment has grown highly popular due to its low cost and ease of use, the possibilities that the electrospinning method allows are nearly unlimited. In this review, the recent applications of metal/metal doped electrospun membranes have been reviewed in a concise manner.

Nanoengineering of metal/metal doped electrospun nanofibers has introduced a slew of new micro- and nanostructural features and behaviors. By creating surface nano-topography or multi-channel fiber structures, hierarchical structures can be incorporated into electrospun nanofibers, increasing their surface area and allowing the incorporation of non-spinnable metals/metal nanoparticles or metals/metal oxides with volatile functional properties during electrospinning. Electrospun nanofibers can be functionalized to improve and alter their physicochemical properties. Apart from doped metals/metal oxide nanoparticles, a variety of agents can boost catalytic activity by immobilizing on nanofibers, allowing for versatility and the potential for a wide range of catalytic applications. Metal/metal oxide nanoparticles doped electrospun fibers will focus on developing increasingly complex fibrous structures and geometries with highly precise control and reproducibility, as well as sustainability, in the future. However, for industrial-scale synthesis and commercialization of electrospun nanofibers in a wide range of catalytic applications, significant modifications to existing or innovative processing processes are required. More crucially, the recyclability of these utilized metal/metal oxide doped nanofiber membranes, as well as the performance boost it can bring, should become a top priority in all application sectors, including catalysis. The use of recycled metal/metal oxide doped nanofiber membranes will be a hot topic in the future since it has the potential to be extremely useful.

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