A Comprehensive Review on Applications of Graphene in Water Purification

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Abstract— Developing the graphene-based membranes is gaining importance for use in wide range of applications including wastewater treatment, desalination, separation, and purification because of its unique properties of high mechanical strength, hydrophilic property, superior flexibility, simple processing, atomic thickness, and compatibility with other materials. In water purification basically high separation performance as well as stable graphene based laminar structures were pursued. Graphene oxide has provided desirable performance to purify water due to its unique water transport property, GO types, structure, properties, membranes, mechanisms, pollutants removed are main point of focus. The characterization of graphene-based membranes by x ray crystallography, UV-VIS spectroscopy, FTIR and other techniques are discussed.

Index Terms— Adsorption, desalination, filtration mechanism, GO membrane, magnetic graphene oxide, nonporous graphene membrane.

1 Introduction

THE essential resource for all life on earth is fresh water, but fresh water is only 0.5% of the earth overall sources as compared to sea water 97% [1]. The available supplies of freshwater are decreasing due to rapid population growth of global population, global climate change, urbanization, rapid industrialization, and more stringent health-based water qualities [2]. According to the report by united nation world water development around 748 million people do not have access to pure drinking water. According to world health organization in 2002 stated deficiency of clean and safe water accounted for 3.1% death worldwide [3]. Our world is dealing with extreme challenges in meeting rising demands of clean water. Across the globe, the increasing scarcity of freshwater sources has urged the need for the development of alternative water supplies that includes seawater desalination, reuse and recycling of wastewater and storm water. [4][5] The most abundant available source on earth for drinking water and industrial use is sea water but it cannot be used for domestic purposes due to its high salinity. Desalination is the process of removal of salt from sea water and making it applicable for drinking. [6] With the increase in population (estimated population increase from 7-10 billion by 2050) and growth of industrialization. organic pollutants, industrial, agriculture and domestic waste that causes water pollution. These kinds of contaminants present in water are very harmful for human beings and for the aquatic life, because of accumulation of these contaminants leads to high risk of health problems for humans as all the contaminants enter the human body through water consumption and cause diseases in humans that include human hepatic dysfunction, carcinogens hindering the development of human body and endangering human endocrine system Water pollution is problem of entire world. figure 1 a) shows types of water pollution. Water pollution is increasing day by day, one of the main causes of water pollution is.

So, water purification needs to be performed to remove the contaminants from water and making sea water drinkable by performing desalination process, various techniques have been used but all these techniques are quite expensive and not that efficient [7].

1.1. Membrane based separation technology

In the past decades, membranes-based separation technology has gained value in water purification due to its positive impact on environment, energy efficiency and easy use. We can categorize membrane-based desalination process on the basis of pore size of membrane and rejection mechanism as: Membrane distillation (MD), Electro dialysis (ED), Microfiltration (MD), Ultra filtration (UF), Nano filtration (NF), Reverse osmosis (RO) [8].

Reverse osmosis (RO) is one of the membrane-based desalination techniques used instead of thermal desalination methods including multistage flash and multiple effect distillation because these methods are not

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energy efficient. The energy consumption of RO has decreased from 5KWh/m^2 in 1990s to 1.8KWh/m^2 today, less than other techniques. However, the RO technique has very low desalination capacity and high capital loss [9].

1.2. Challenges for water desalination membranes

One of the primary concerns in desalination technology is membrane fouling, due to which RO membrane performance declined [10][11]. The membrane fouling occurs by one of the two mechanisms: fouling in membrane pores, membrane surface fouling. The membrane surface fouling mainly occurs due to presence of impurities including biogenic materials, suspended inorganic or organic matter and dissolved solids in feed water [12]. The major limiting factor in sea water desalination process [13] is particularly the biological impurities which build up on membrane surface during RO process and forms biofilms [14]. the growing biofilms is removed by using disinfectant such as chorine but they can react with PA on membrane surface layer, it is found evident by research studies on RO membranes [15]–[18]. The reaction of chlorine with PA causing variations in capacity of thin film composite of RO membrane(TFC) [19], even if we use low concentrations of chlorine.

The polymeric membranes used in RO suffer from flux decline under high pressure, low tolerance to chlorine, acids and alkalis and high temperature so there is need to develop energy efficient membranes [20].These polymeric membranes are being replaced by carbon based materials, nanostructures such as zeolites, ceramics which are getting attention due to their high rejection rates, high flux, good chemical resistance [21].But these materials also have certain limitations as carbon nanotubes have tendency to fabricate high flux and high selectivity membranes but high cost and operational issues have prevented its study and making it difficult to develop CNT into large area membranes [22]. Zeolite membranes have also failed because it is not economical to fabricate it on large scale due to its reproducibility, cost, and defect formation. [23] While ceramic membranes have also limited practical applications in membrane technology because they are very costly and brittle under high pressure.

Graphene based materials are gaining great interest in recent times for their potential exploitation in water desalination and purification membranes. The studies are done on graphene and advancement in molecular simulation of graphene family has opened the door for their contribution in developing novel membrane desalination techniques.

1.3. Graphene

Graphene was first mechanically exfoliated from the graphite by simple scotch tape method by DR. KONSTANTIN NOVOSELOV [24] and professor ANDRE GEIM [25] in 2004. The building block of graphite is graphene, the graphite structure has sheets of graphene stacked together with interlayer spacing of 3.34 angstrom [26]. Graphene can be defined as 2D sheets that are one atom thick; they consist of sp^2 bonded carbon atoms and are arranged in hexagonal, honeycomb lattice. Carbon atoms in graphene form four bonds three sigma and one pi bond, sigma bonds are formed with three neighboring atoms and pi bond is oriented out of plane. It has large surface area (2630 m^2 g^{-1}), high absorption capacity, high electrical conductivity even 13 times better that of copper (~5000 Wm^{-1}k^{-1}), high thermal conductivity 2 times more better than diamond and attracted attention for almost 40 years [27]

One of the most important characteristic that makes it so important is its extremely versatile carbon backbone that leads to facile functionalization and incorporation in variety of applications and it can easily be fabricated to be used on large scale as recent work is done on production of 30 inch multilayer graphene sheets and being transferred on roll-to-roll fabrication [28]. Graphene and CNTs have ability to inhibit bacterial growth by direct contact with cells. Figure 1 c) shows Graphene based materials are used in wide range of applications but we are focusing on water purification.

Graphene based materials include: Graphene oxide (GO),Reduced graphene oxide (rGO),Graphite (Gt),Graphite oxide (GtO),magnetic graphene oxide(MGO) By comparing different graphene based materials in terms of their antibacterial activity against Escherichia coli (bacterial species) .At the same concentration, incubation time and conditions, GO dispersion has the most antibacterial ability than others so, Figure 1 b) explains Instead of using graphene alone it's better to use graphene oxide that has better oxidative properties than graphene or Nano porous graphene (NPG).[29], [30]

1.4. Nonporous graphene membrane

NPG are also potential candidates to excel due to following characteristics. For introducing Nano pores into graphene, various experimental procedures have been explained and major developments achieved. In graphene the sub-nanometer sized pore can be defined with high precision by using methods like ion bombardment and oxidative etching methods [31], [32]. It has possibility of achieving 100% salt rejection. By managing the porosity on graphene, the water permeation rates can be governed.

NPG can provide high water flow rates and high salt rejection as function of Nano pore morphology but single layer of graphene sheets are difficult to assemble so there remains a major challenge to achieve scalable manufacturing of large NPG membranes and cost effectives having a required pore size and narrow size distribution while preserving intrinsic structural integrity of graphene [23].The demerits are associated with NPG for desalination as it is extremely difficult challenge to achieve narrow size distribution of holes with high density and also these tiny holes generated in graphene can restrain the amount of needs extreme cautions, even if we achieve high water permeability. The high-density holes can cause reduction in mechanical properties or destruction of whole structure. Lastly, the oxidative etching technique, design of high quality graphene and ion bombardment are quite expensive methods [33]. The most recent applications are in gas separation and DNA sequencing.

2.GRAPHENE OXIDE (GO)

GO was first developed by **Nair et al** [34] as a feasible membrane and he discovered that by stacking graphene oxide film it allows unique water permeation pathway, but it selectively hinders the motion of gases and non-aqueous solutions. Due to 2D structure, one atom thickness and excellent mechanical strength and flexibility the graphene-based materials mainly graphene oxide has been widely studied for the thin film and membrane applications [35]. For the development of ultra-thin and high-perm selective membranes, GO has gained importance because GO has great number of functional groups like OH, COOH etc., high area to thickness ratio and becomes excellent material for membrane fabrication. GO fragments can easily be arranged on top of each other to give a membrane [36], [37]. It is permeable to water flow but does not allow vapors to pass through it [38] .As nothing can pass through graphene but super membrane properties are shown by GO and it is used commercially.

The advancement of GO and reduced form of graphene has made them applicable for water purification purpose[36], [39], [40]. The capillary driving force and low frictional force between the two 2D channels of graphene results in high permeability that is far better than that of other membranes used for purification.[39]

2.1. Preparation of GO

Graphene oxide (GO) has more stretching ability because of –OH, -COOH and C-O-C functional group presence. GO has imperishable hydrophilicity. It has more application than graphene. Graphene oxide can be synthesized mainly two steps: figure 1 g)

→ By oxidation of graphite. → By exfoliation of graphite oxide, exfoliation is the process of transforming the stacked layers of graphite into single layers of graphene.

GO membranes are used for water purification but can be prepared (used) by various methods including **Brodie**, **Staudenmaier** [41] or **Hummer** method [32], [42]–[44] figure 1 e). The first two methods oxidize graphite by using nitric acid (HNO_3) and potassium chlorate ($KClO_3$).[45]

Hummer method provided rapid, safe, and more capable method for forming graphite oxide before this method the fabrication of GO was unsafe due to use of intense nitric acid and sulfuric acid. Han et al [46] succeeded in fabricating GO via modified Hummer method. This

water permeation, the whole process of composing NPG

method oxidizes graphite by using combination of $KMnO_4$ and H_2SO_4. The salts prepared by intercalating graphite with acids i.e., sulphuric acid (H_2SO_4), nitric acid (HNO_3 which are created prior and then exfoliation step is carried out.

GO based membranes can be used in following way:

2.1.1 ***Free standing (GO directly used for separation):***

Xu et al reported continuous vacuum filtration technique that can be applied to fabricate free stand GO membrane with gaps among GO nanosheets [50]. GO/TiO_2 composite nano filtration membrane were fabricated with pore size having average of 3.5nm. they introduced TiO_2 nanoparticles between the nanosheets so to widen the gap and forming channels. This NF GO/TiO_2 membrane has achieved rejection of contaminants to about 100% from water. GO/TiO_2 sheets were stacked into well-packed alignments of GO/TiO_2 membranes. These TiO_2 nanoparticles could support GO Nano sheets and enlarge the interlayer Spacing, which result is the formation of pores and suitable channels in the fabricated membranes and allowed them to be promising filtration membranes for water pollutants.

Nair et al [47] minimized the inetersheet gap to about 1nm and his researches include fabricating free standing Cu-assisted GO Nano filtration membrane by spray or spin coating method, the resulting membranes were completely impermeable to gases, vapors and liquids as well as helium gas and permitted water flow. Moreover, the rate of water permeates were 10 times faster than that of helium.

Sun et al [48], by using drop casting techniques fabricated GO freestanding NF membranes and intersheet spacing reduced to 0.82 nm and these membranes can easily separate sodium salt from copper salt and organic contaminants. Freestanding GO membranes have an exceptional ability of rejecting salt by 100% and have high water permeation and have permeation ability almost double to current RO techniques because of their ultrathin thickness (~10nm).

2.1.2 ***Supported (by polymeric or inorganic substrate):***

If the GO is properly combined with ceramic, polymer matrices or substrate, the properties could be improved.

The GO incorporated membrane are used in various application including: Fuel cells [48][49] Nano filtration [50], [51]

Ultra-filtration [52], [53] Gas separation [54], Pervaporation GO can enhance the performance of obtained hybrid polymer membrane including its antifouling, mechanical and surface charge properties. The hydrophilic nature of some functional groups in GO are reason of improvement of GO membranes and causes better GO dispersion in water and organic solvents forms mesh as in nano size GO

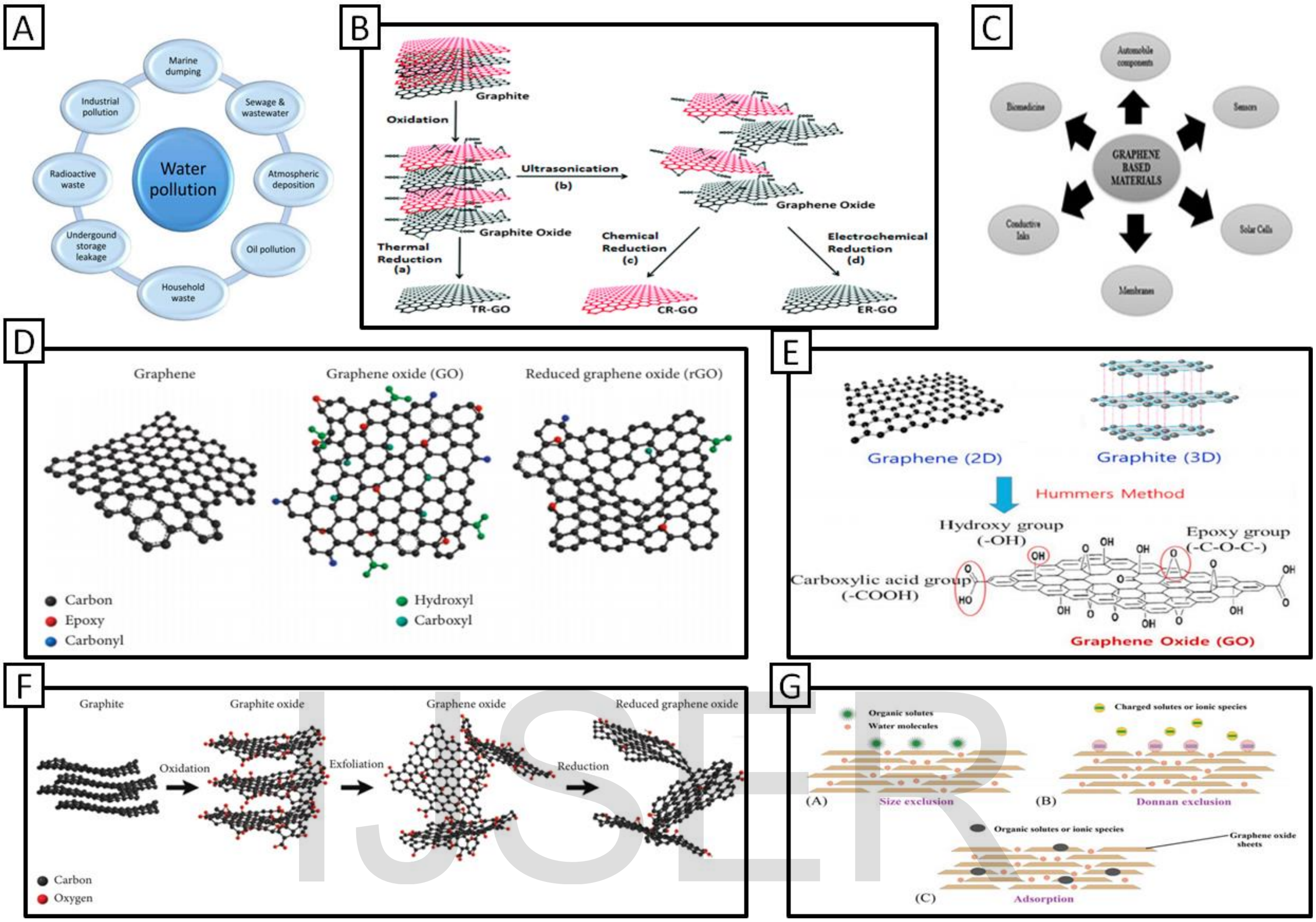

Fig 1.a) Types of water pollution (reproduced from Copyright © 2021 Elsevier B.V.) **b)** Types of graphene-based materials (reproduced from Copyright © 2021 Elsevier B.V.) **c)** Different conversions of graphite, graphene oxide Adapted from, with permission from ©2012 Royal Society of Chemistry **d)** Structures of graphene, graphene oxide and reduced graphene oxide (Copyright © 2019 P. Jayakaran et al **e)** Preparation of graphene oxide (reproduced from Copyright © 2021 Elsevier B.V.) **f)** Oxidation, exfoliation, and reduction of graphite (Copyright © 2019 P. Jayakaran et al **g)** Filtration mechanism (@2019 Elsevier Inc. All rights reserved.

across hydrophobic carbon core and developing water channels that improve permeation flux. Water molecules when infiltrate GO layers, they create a single layer configuration that will drive the other consecutive layers apart, resulting in increased d spacing.

2.1.3 Graphene oxide membranes supported on the ceramic hollow fibers for efficient H_2 recovery:

For the recovery of $\mathbf{H_2}$ from gas mixtures, oxygen-nitrogen separation, natural gas separation, CO_2 separation and air dehydration, the gas separation membranes are gaining importance [55].This is because the membrane technology can more efficiently separate gas mixtures under low pressure, minimum energy consumption with low contaminants as compared to traditional separation technologies [56] GO membranes are most suitable for gas separation because of their precise transport channels and atomic scale pores [57] But there are certain shortages including membrane preparation process [58].

So, it is difficult to use these membranes for practical applications. A careful manipulation is required for these few layers GO membranes. These shortages can be overcome by preparing GO membranes supported by porous ceramics because these substrate offer good mechanical strength for composite membranes [59] and decreasing thickness of membranes to realize high flux. A vacuum suction method is used to prepare GO membranes on ceramic hollow fiber which can exhibit excellent separation performance [60]. The permeances of small gas molecules like hydrogen, carbon dioxide, oxygen, nitrogen, and methane decrease with increasing molecular weight.

2.1.4 Substrate effect:

The bulk pore structure of substrate as well as surface morphological and chemical structure of substrate effect the interfacial adhesion and Nano filtration performance of GO membrane.

In PAN [61] due to the presence of abundant oxidized functional group leads to enhanced interfacial adhesion of GO membrane and highly porous sublayer with flat skin allows very low transport resistance as a result excellent nanofiltration performance of GO membrane. The PAN substrate has a pore size of 20 nm, consists of very thin, dense top layer roughly 1micrometer thick, it has finger like cylindrical pore structure bottom with large tubular macro voids. These features provide a flat platform for assembly of GO laminates and large channels for water permeation. This PAN substrate generated water permeance of about 585 $Lm^{-2}bar^{-1}$ which is more than 5-fold more than that of ceramic substrate.

PC [62] substrate owing to its circular straight through pores possess ultrahigh water permeance but has a very poor adhesion performance due to lack of surface functionalities. When PC substrate has pore size of 200nm with ultrahigh water permeance of 4575 $Lm^{-2}bar^{-1}$because of low transport resistance, it exhibits straight pore channel with no tortuosity through its bulk structure, but internal pore channels of PC get easily collapsed and cause deformation and even crack of bulk pore structure.

2.1.5. *Modified composite membrane (by directly incorporating GO Nano sheets into polymer casting solution during membrane fabrication process)*

Recently research have been done, in order to improve water permeability, mechanical strength and antimicrobial properties. GO must be mixed with polymers and casted together. Polymer solutions like polyvinylidene fluoride (PVDF) and polysulfone (PSF) mixed with functionalized GO (f-GO) to be casted together by using common phase inversion method.

GO enhanced polyamide (PA) thin film nanocomposite (TFN) membrane for water purification purpose:

In membrane separation area ultra-thin membrane of graphene is used to separate gases present in the water so graphene nanosheets or graphene nanocomposite are used because of their potential application for purification of water for better separation graphene is enhanced by attaching its derivatives like GO and reduced graphene oxide on polymeric substrate this resulting membrane shows more efficient results than that of graphene. Through vacuum filtration of a reduced graphene oxide, ultrathin graphene nanofiltration (NF) membrane on top of which microfiltration membrane is placed. Similarly, TiO_2-graphene oxide membrane can be prepared by this method.

Another way includes GO nanosheets into the polymeric matrix. Although graphene is not suitable with organic polymers, likewise, GO sheets having hydroxyl, epoxide, diol, ketone can change the interaction of GO sheets and polymer matrix. GO nanosheets can be prepared by hummer method and then fix into polyamide thin film layer by interfacial polymerization. GO nanosheets with many layers structure was prepared and used as the filters to fabricate the TFN membrane so nanosheets were dispersed good in PA thin film. As the concentration of nanosheets increases hydrophilicity of the TFN membrane also increases, similarly water flux is also increased.

Amorphous feature of the membrane can be increased by mixing the functionalized hydrophilic nanoparticles with the polymeric matrix. Through the rapid exchange between the solvent and non-solvent during phase inversion procedure, the porosity of f-GO was enhanced) (dense pores the introduction of f-GO improves the pore size, porosity but it decreases by the further additives incorporation

2.2. Filtration mechanism

GO membranes having the thickness of submicrometric can completely hinder liquid and gas molecules flow and allowing the permeation of water vapors only [63].this may be due to the presence of empty space between the non-oxidizing sheets of graphene oxide, that is permeable to water vapors through low friction channels. The possible filtration mechanism of GO membranes are as follows:

- Size exclusion
- Donnan exclusion
- Adsorption phenomenon

Because of presence of Nano channel in membrane the bulky organic molecule can be filtered out. For the precise separation of bulky molecules and ionic species, the size of Nano channel has to be increased by manipulating spacing between graphene oxide sheets [64] for desired application like desalination, water purification and pharmaceutical we can fabricate graphene oxide membrane with specific spacing in layers. we can also use **Donnan exclusion process** [65]In this process, negatively charged organic species or divalent ions can be rejected by pure graphene oxide membrane because GO membrane gets negatively charged as the proton from.

2.2.1. ***Direct contact membrane distillation**:*

To provide cost effective alternatives to desalination, distillation and reverse osmosis are of great value. Membrane distillation [66] can be defined as thermal evaporation process (based on membrane) that can be done at low temperatures of 50 to 90 degree centigrade and resulting in energy-efficient water generation. In MD between hot feed and cold permeate of hydrophobic porous membrane distillation by immobilization of graphene oxide on PTFE this enhances the result rejection ability of membrane and higher flux of 97 kilogram per meter square or at 80 degree centigrade. Multiple factors are responsible

- nano capillary effect
- reduce temperature polarization.
- polar functional groups in graphene oxide (figure 2 a)

Graphene oxide is gaining importance in MD. however, graphene oxide real world application remain at challenge as a single sheet of graphene are difficult to assemble. In this we are basically discussing desalination via a direct contact selective sorption.

Graphene has two dimensional sheets in which sp_2 bonded carbon atom is hexagonal in structure they acts as a sorption site for water vapors that are formed from hydrogen bonding the saltwater clusters are repelled by the PVDF .The flux can be enhanced by preferentially hydroxylating or carboxylating the carbon atoms adjacent to pore so the main objective of the research is to immobilized graphene oxide on PTEE membrane to fabricate high performance desalination membrane for MD. the overall water permeation rates for enhanced due to the presence of polar functional groups like epoxy etc. Sodium chloride, PVDF powder, cyclohexanone and DI water are used in all experiments. The graphene oxide single atom layer the carbonyl group is removed that is present on the edge or tape of graphene oxide sheet figure 1 h).

Another mechanism is **adsorption phenomenon.** Small ionic species can be rejected by GO membrane by strung adsorption via interaction with varying regions of graphene oxide sheets. This is because of fact that oxygen containing functional groups of graphene oxide membrane [67] form co-ordinate with transition metal cations resulting in complete blockage and in same way alkaline earth metals and alkali permeability also lessened due to their interaction with sp2 plaster of graphene oxide sheets via Pi interactions [68] membrane act as a barrier. As it allowed the vapors to pass through the pores so be condensed on permeate side but does not allow passage of liquid phase. Here the key to flux and selectivity is membrane itself. By improving membrane architecture, we can enhance MD performance. graphene oxide immobilized membrane on the permeate side (GOIM-P) membrane with 0.2 micrometer per and 70% purity and 35 micrometer thick PTFE membranes with non-woven polypropylene support were used.

3.REDUCED GRAPHENE OXIDE

We must reduce graphene oxide membrane because of d-spacing could be decreased due to the presence of hydrophilic functional groups on the edges of surface of graphene oxide and reduction remove these functional groups. The reduced graphene oxide is prepared either by

- chemical reduction
- thermal annealing

for this enhancement in flux.

To remove oxygen based functional groups like OH, Most effective method for thermal annealing [69] is the thermal deoxygenation of graphene oxide is assisted by a rise in temperature but in this process consumption of energy is high and degree of oxidation is very difficult to control. chemical reduction requires low temperature range by adding reducing agent such as metal hydrides, hydrazine, hydroiodic acid the target of these functional groups is difficult. Reduced graphene oxide membranes based nanofiltration membranes have better properties than graphene oxide, so number of research have been done on them. Graphene based nanofiltration membranes (NF) was developed by depositing the magnesium silicate modified reduced Graphene oxide (MgSi@RGO) nanosheets on a polyacrylonitrile (PAN) by filtration method.

- *Synthesis of MgSi@RGO nanocomposite*

Modified hummers method was used to prepare GO samples. The resulting GO was centrifugated to get a homogenous suspension. After dilution MgSi@RGO was prepared Silica/RGO is allowed to react with MgCl, NH_3Cl and NH_3H_2O by hydrothermal process.at the end the resulting outcome was then centrifugal rated washed and dried.

- *Preparation ofMgSi@RGO aqueous suspension*

To get a homogeneous aqueous suspension, the prepared MgSi@RGO samples was again dispersed in deionized water. After this resulting dispersion was sonicated for some time.

- *Preparation of MgSi@RGO/PAN NF composite membrane*

PAN membrane was dipped into an ah solution by hydrolysis process the MgSi@RGO skin layer was prepared by filtration and dilute MgSi@RGO aqueous dispersion was filtered the MgSi@RGO/PAN NF composite membrane consists of thin MgSi@RGO layer and substrate ultrafiltration membrane. Then dried in vacuum before use.

- *Synthesis of r GO-TH and r GO-TH membrane*

rGO membranes with enlarged interlayer distance using graphene oxide sheets and tannic acid (TA) and the amine amino acid (TH) as reducing agent have been synthesized. These membranes have Ultra high-water permeability and excellent stability membrane show 10-1000 times greater durability than those of previously reported graphene-based membranes. Moreover, they show no damage in acid basic solution even after months. Figure 2 b GO sheets were prepared by exfoliation of graphene oxide made by hummers method; they are monolayer with thickness of 1nm. To fabricate rGO-TH membranes some amounts of GO & TH were weight with ratio1:1 first dissolved in DI water then stirred for overnight at 70 degrees. Color changes from brown to black indicating reduction of graphene oxide sheets. The thickness of membrane was controlled by content of GO & TH. We have prepared graphene oxide-based membrane with best efficiency by using graphene oxide sheets and TA/TH molecules. It is found that they play four roles:

1. link reduced graphene oxide sheets as crosslinker
2. enlarge reduced graphene oxide interlayer distance.
3. block the solute together.
4. reduced graphene oxide sheets to increase Pristine graphene.

4. MAGNETIC GRAPHENE OXIDE (MGO):

Graphene oxide has high dispersibility, so its separation from the aqueous solution even after adsorption of pollutants is quite difficult. This problem can be avoided by magnetization of GO. The magnetized GO can easily be separated by external magnetic field. Moreover, the magnetic particles show high adsorption capability towards pollutants, so they are coupled with GO. Several MGO has been synthesized by incorporating metal oxides in GO based nano composites including: Fe_3O_4/GO, r MGO, Mn_3O_4/GO, nickel ferrites and other hybrid nano composites. Nickel ferrites are better reaction media then that of iron ferrites because they have higher catalytic and charge transfer efficiency through Ni^{+2}.

4.1 Applications to sustainable water purification:

GO has substantial hydrophilic groups and possess a large surface area and it is a good adsorbent. GO exhibits more hydrophilic nature than graphene because it has oxygen containing functional group and forms more stable complex with pollutants and it get easily dispersed in solution but GO faces an issue of its recovery which can be solved by doing magnetic functionalization of GO and used for the removal of pollutants. Magnetic graphene oxide composites have found applications in water treatment, energy storage and drug delivery. Figure 2 c)

4.1.1 Agricultural pollutants:

Pesticides:

Due to excessive use of pesticides for killing insects, they get accumulated, and their residues contaminate soil, crops, vegetables and is not good for human health [82]. these pesticides include neonicotinoid insecticides such as dinotefuran, clothianidin, imidacloprid etc. Liu et al [83] synthesized magnetic copper-based metal organic framework using Fe_4O_3-GO nanocomposite as support and used for the adsorption and these insecticides get easily separated.

Herbicides:

Atrazine is weed killer or herbicide; it is used in the crops such as maize and sugarcane to prevent pre and post emergence of weeds but nowadays it is used as turps such as residential lawns and golf courses to eliminate weed but theses may enter water body through rain water and it has adverse effect on amphibian sexual development and also lead to reproductive effect in humans. For adsorption of atrazine from aqueous solution. Liu et al [70] synthesized magnetic molecularly imprinted polymer (MMID) using g-C_3H_4-Fe_3O_4 as a support. Another study by Boruah et al [71] prepared Fe_3O_4 supported rGO nanocomposite for remiving harmful pesticides naming simazine, simeton, ametryn.

Opioids:

Opioids (methandone) are generally used to treat pain and sold in market as dolophine, as maintenance therapy or to treat people from opioid dependence. This is found in very trace amounts in wastewater due to unlawful medications. This methadone can be adsorbed from the environment by means of MGO [70]

4.1.2. Organic pollutant removal:

Radioactive metal ion removal:

The radioactive waste released from nuclear energy production activities and mining operations has a long-term effect on environment. Radionucleids such as Sr^{90}, Cs^{137}, U^{235}, I^{129} released as a product in fission process can enter food chain by water systems and water contaminated by these radionucleids may sneak into soils and adsorbed by plants and ultimately reaching animals and human beings. Other sources of radionucleids are ore processing, lignite burning in power station and use of fertilizers. Major concern is contamination of freshwater by these radionucleids.

Zhao et al [74] worked on it, GO based nanosheets are not used because of their aggregation so functionalization of GO by magnetic materials was considered as a effective method. Sun et al synthesized iron-r GO (nanosized zero valent) for the adsorption of U(VI).

Nickel ferrite-GO nanocomposites synthesized by Lingamdinne et al [75] can be used for the treatment of U(VI) and Th(IV) , MGO nanocomposites have high adsorption capacity for both compounds, these nanocomposites can be reused up to five cycles.

ZnO:

Graphene based nanoparticles are used for the decontamination of sea water. These include graphene oxide, reduced graphene oxide and graphene coupled with ZnO. These materials received a lot of attention due to excellent qualities like chemical stability, non-toxicity, excellent electrical conductor, large surface area, good mechanical strength, stiffness and long range of porosity.

In recent times advanced techniques have emerged as a unique and efficient approach for the decontamination of organic as well as inorganic pollutants present in water.

Advanced oxidation processes included the reactivity of OH radicals in the oxidation process which includes the series of reactions and degrade the contaminants into

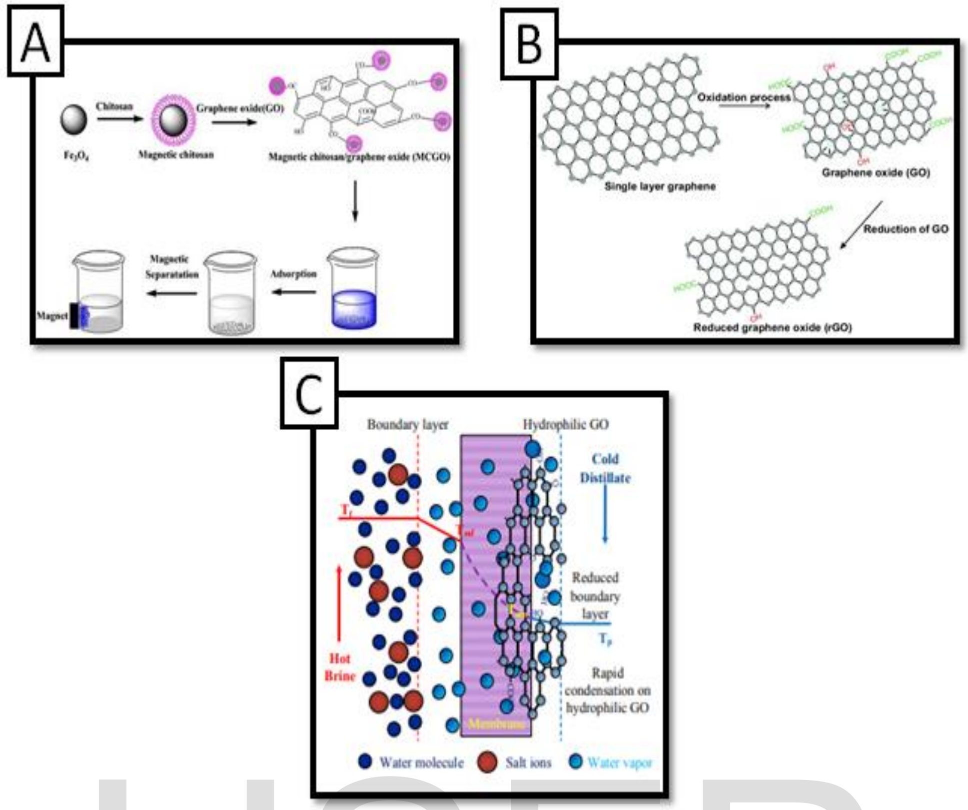

Fig 2. a) Magnetic graphene oxide (MGO) (@ 2012 Elsevier Ltd. All rights reserved) **b)** Synthesis of reduced graphene oxide (Copyright © 2018 Elsevier Inc. All rights reserved) **c)** GOIM-P Proposed mechanism (© 1996-2021 MDPI)

offensive product.

Numerous metal oxides are used for the water purification like nibium pentaoxide (Nb_2O_3), titanium oxide (TiO_2), iron oxide (Fe_2O_3) etc. ZnO nanocomposites [80] have a large surface area and more number of active sites for the absorption of contaminants from water. ZnO nanoparticles is manufactured by the saturation of graphene sheets over it by simple solvothermal process.

5.CHARACTERIZATION:

GO membrane produced can be physically characterized to ensure the characteristics before using membrane for filtration. Initially to confirm the surface morphology and laminated structure of GO membrane, the scanning electron microscope (SEM) was used.

X ray diffraction technique was used to determine the interlayer spacing between laminates in the membrane and to show changes in crystallographic structures. The sharp peak is obtained at 2 theta value of 10.7 degree that is equal to d spacing value of 8.25 angstrom for membrane that is typical for GO. The peaks of MgSi@RGO and MgSi sample are broadened and overlapped with each other, but the peak of RGO was too waek to be detected or overlapped with MgSi peaks so difficult to calculate spacing brGO was fully characterized by atomic force microscopy (AFM), thermogravimetric analysis (TGA), raman spectroscopy and x ray diffraction (XRD) analysis. The brGO sheets showed uniform lateral size around 1-2mm and performed single layered feature deposited on mica with 0.8nm thickness.

Two main peaks at 12.6 and 24.1 degree are observed in XRD [100] pattern of brGO in powder state with spacing approximately 0.7 and 0.4nm. Double peaks show certain number of functional groups still on brGO. Notes: Dried powders of gO, rgO, and rgO–silver nanoparticle (agNP) nanocomposite were diluted with KBr to perform FTIr spectroscopy and spectrum gX spectrometry within the range of 500–4,000 cm −1.

We can also use **FTIR** spectrum that indicates presence of OH stretching vibration ($3415cm^{-1}$), carbonyl group stretching vibrations ($1733cm^{-1}$), unoxidixed sp^2 carbon double bond carbon bonds ($1624cm^{-1}$), carbon single bond oxygen from epoxy groups ($1051cm^{-1}$).

Notes: Dried powders of GO, rGO, and rGO–silver nanoparticle (AgNP) nanocomposite were diluted with KBr to perform FTIR spectroscopy and spectrum within the range of 500–4,000 cm −1.

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XPS (x ray) photoelectron spectroscopy can also be used for

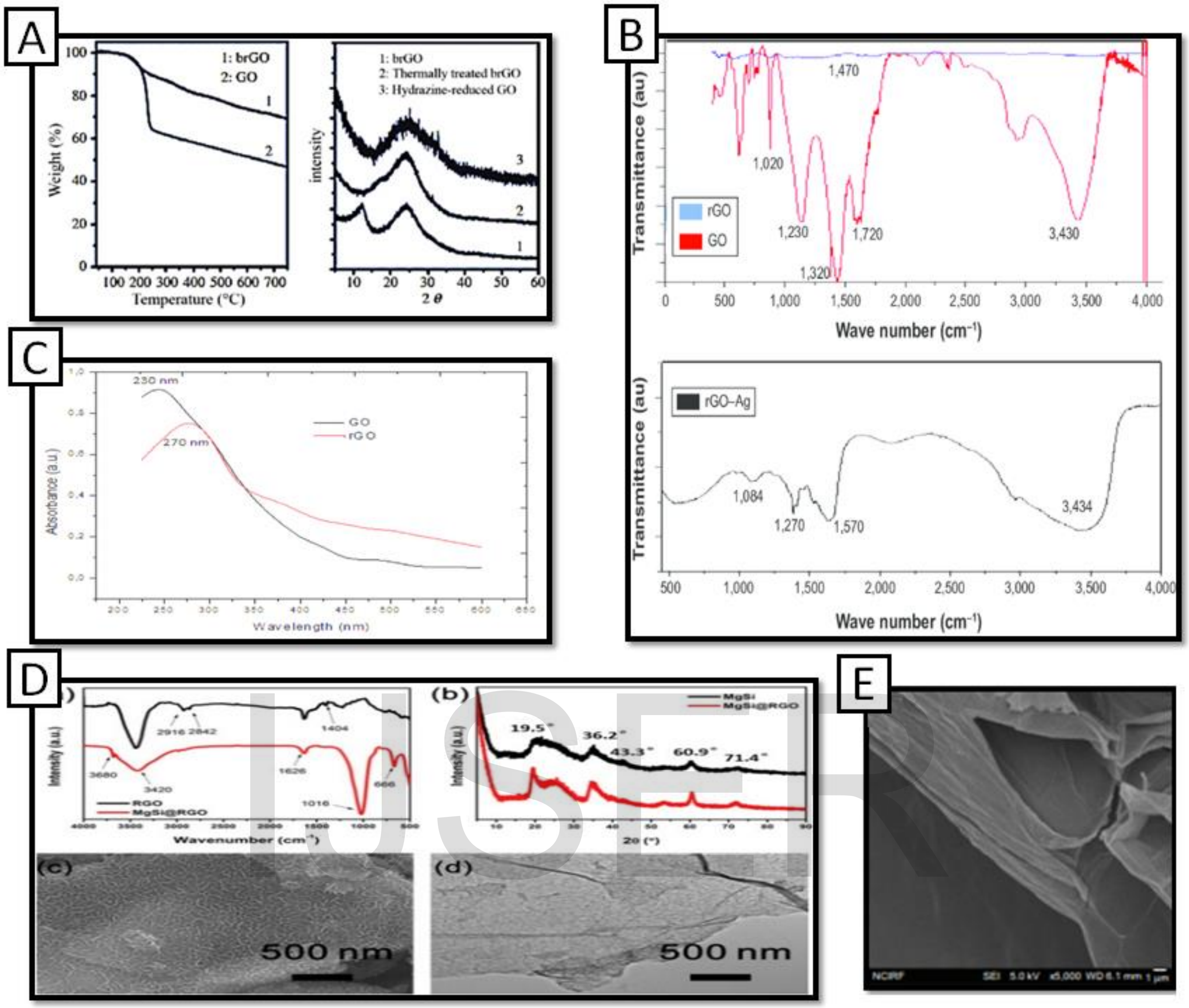

Fig 3 a). a) Thermogravimetric analysis in N of GO and BrGO (b) XRD pattern of GO and BrGO Copyright © 1999-2021 John Wiley & Sons, Inc. All rights reserved. **b).** a) Fourier transform infrared (FTIR) spectra of graphene oxide (GO), reduced graphene oxide (rGO), b) rGO–Ag nanocomposite. © Copyright 2021 Dove Press Ltd **c)** UV-Vis spectra of GO (graphene oxide) and rGO (reduced graphene oxide). (1995–2020 Copyright Clearance Center, Inc. All rights reserved) **d)** MgSi@RGO and MgSi XRD pattern is shown below in (a) and (b)(c) FESEM images of MgSi@RGO nanocomposite; (d) TEM images of MgSi@RGO nanocomposite. (Copyright © 2021 Elsevier) **e)** SEM image of GO(© Copyright 2021 Dove Press Ltd)

characterization, it indicates four kinds of carbon atoms in different functional groups C-C (~284.8ev) , C-O (~286.8ev), C=O (~287.8ev) and C(O)O (~289.0ev).

6.CHALLENGES

There are many commercial applications of graphene but so far, the real application in industrial or commercial products has been very limited. This wonder material has not been embraced by industries and other companies for many reasons: At this moment 1g of graphene is estimated to cost roughly US$100 and with this cost commercialization takes years and it is very difficult to produce graphene in large amounts with no defects, but still there are companies which are working on it to make it better.

A lot of research work and remarkable discoveries have been made in the field of graphene and graphene-based materials study, but despite all this several challenges exist in the commercial application of graphene oxide membrane.

- Major issue is membrane's stability graphene oxide membrane have shown in ability in air as after the drawing process their size reduces, which induces in stability and work was done to avoid this reduction issue that can be solved by storing membranes in water medium so we can avoid

- excessive drying [72], [81] or during membrane preparation employing upper sacrificial coating. [82]
- Recent work on instability improvement of graphene oxide membrane has depicted that by using ceramic material has a substrate improves the stability of membrane in aqueous medium by releasing various multivalent and ions that allow cross linking of graphene oxide sheets, but still further work is required for resulting this stability issue when membrane with it just at submicron level. There are also limitation on characterization of graphene oxide membrane in several studies.[40], [83]

7.CONCLUSION

Graphene and graphene oxide has been significantly used to improve membrane properties for desalination and organic pollutant removal applications. GO after incorporating into membrane has improved properties like antimicrobial, antifouling, mechanical strength, selectivity, water flux and thermal properties. To remove organic pollutants photo catalysts like Zn and titanium oxide can be used combined with graphene. Graphene oxide membranes can help in removal of heavy metals, removal of dyes, separation of monovalent and divalent ions. Various fabrication techniques have been developed. The GO nanosheet is the most important building block for the fabrication of GO assisted membranes but more attention should be given to their drawbacks like stability issues. On of the greatest challenges faced today is scaling up the commercial fabrication of ultrathin GO membrane, we have to strike balance between costs and the simplicity in manufacturing operations. GO membranes could be one of the most important tools to solve the expected world crises of water in near future. [84]

8. END SECTIONS

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8.2 Reference

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