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Article

A Review of Polymeric-Inorganic Composite Membranes: Characteristics, Classifications, and Its Applications in Water Purification Process

Mohd Arsalan*

Department of Applied Chemistry, Aligarh Muslim University, Aligarh, Uttar Pradesh, India

ABSTRACT

Membrane is a thin physical wall which can be defined as an interface between two adjacent phases functioning as a selective barrier and regulating the transportation between them. A thin sheet of natural or synthetic material that is permeable to substances in solution depends on their physical or chemical properties. Polymeric-inorganic composite membranes are now one of the most important classes of engineered materials which offer several outstanding properties as compared to conventional membranes. They can be produced from polymers such as PVC, Polystyrene, and PVA and inorganic materials such as metal phosphate, tungstate, arsenate, and many metallic oxides are also used to make the synthetic membrane. The most of commercially utilized synthetic membranes in separation industries are made of polymeric and inorganic composites. They can be classified, based on their surface chemistry, bulk structure, morphology, and the production method. The chemical and physical properties of synthetic membranes and a choice of driving force define the membrane separation processes. Synthetic membranes can be divided into the categories such as organic (polymeric or liquid), inorganic (ceramic or metallic), composites (organic-inorganic). These composites can be homogeneous films (polymers), heterogeneous solids (polymeric mixes and mixed glasses), and liquids. Homogenous membranes are coherent ion-exchanger gels in the shape of disks, ribbons, etc. The heterogeneous membranes consist of colloidal ion-exchanger particles embedded in an inert binder. Membrane based technologies witnessed much potential to impact multimillion dollar industries in various sectors such as food/drug production, separation of pollutants, water purification, textile, biotechnology and to fuel cell markets.

Key words: Polymeric-inorganic composite, Membrane characteristics, Membrane classification, Membrane applications, Water purification process.

1. INTRODUCTION

A membrane can be defined as an interface between two adjacent phases functioning as a selective barrier and regulating the transportation between them. According to *Sollner* a membrane is a phase or structure interposed between two phases or compartments which prevents gross mass movement between the later, but permits the passage with various degrees of restrictions of one or several species of particles between two adjacent phases or compartments, thereby acting as a physicochemical barrier with varied degree of efficiency depending to the nature and composition of two adjacent phases or compartments [1-3].

The conventional macroscopic composite materials like adobe are a mixture of clay and straw, a composite material used to make bricks and walls in arid regions, and when these are reinforced with concrete, gives structural shaped to our world during past [4-9]. Yet, when it comes to microscopic world, reduced particle size boost the importance of the inter phase in composite mixtures which are especially important for a new class of innovative and developed materials called as micro- and nano-composites materials. To satisfy the demanding applications, it is very essential that the composite membranes must possess high conductivity and selectivity, good mechanical and chemical stability as well as high resistance for the little jerk. To accomplish the above criterion, the selection of polymer (binder) and inorganic compound is very important. Nowadays, most of the composite membranes uses the polymers such as polystyrene, polyvinyl chloride, polyvinyl alcohol, cellulose acetate, polyamide, polysulfone, nylon, polyvinylidene fluoride, poly tetra fluoroethylene, polypropylene, and polycarbonate [10-12]. Those inorganic materials which preferred for the preparation of composite membranes are generally metal phosphate, arsenate, carbonate, tungstate, and metal oxides due to the above-mentioned properties.

The synthesis of polymeric-inorganic composite membranes can be done by several processes such as sol-gel, co precipitation, intercalation, blending, *in situ* polymerization, and molecular self-assembling but among them sol-gel process is one of the most common and qualitative method. Many researchers used the sol-gel or liquid coupling process of silane coupling agents to prepare a hybrid anion exchange membrane [13]. The ionic conductivity of membrane is an important characteristic which is the major deciding factor of application processes. It is due to the type of functional groups such as strong ionic phosphonic, sulfonics acids, and quaternary ammonium salts and the weak ionic such as hydroxyls, carboxylic acids, and primary secondary and tertiary amine groups. It is analyzed that the porosity of membranes majorly affects the ionic conductivity, which suggest

***Corresponding author:**
E-mail: mohdarsalan.chem@gmail.com

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that a highly porous membrane always follow high ionic conductivity and vice versa. Hence, the porosity of composite membrane can easily be modified using the suitable organic and inorganic materials as well as the synthesis ideas [14,15]. Water content property of hybrid membrane has also indicated the effect for ionic conductivity, which means that the high water uptake leads to high ionic conductivity or ionic migration. Hence, the better and appropriate applications in different industrial fields demand the low water content with high ionic conductivity. Therefore, the designing of new composite membrane for the commercial purposes, the required criteria that must be followed is the lower water content as well as higher ionic conductivity.

2. MEMBRANE SEPARATION TECHNOLOGIES

The separation of particles by polymeric-inorganic composite membrane mainly depends on the pore size distribution. If the size of the particles is larger than the pore size of membrane then these are rejected, while the smaller particles can easily pass through the membrane barrier. Hence, the membrane filtration is entirely based on the membranes pore size distributions [16]. The resistance of mass transfer in such type of membranes is totally determined by their thickness and porosity which shows that the membrane thickness is inversely proportional to the permeation rate of transferable particles. Membranes often respond to gradients that they experience on either sides of them. If concentration is a gradient, the dialysis results, if pressure is a gradient then reverse osmosis, ultrafiltration, microfiltration or nanofiltration results, while if potential is a gradient then the electro dialysis and electrophoresis results. All these processes are differing from each other and depend on the pore diameter of the membrane [17,18].

It is possible to exploit various parameters in the development of membranes and among them the electrical conductivity of developed membrane has received a maximum attention. However, in specific applications the species transfer across the membrane can also be another factor of consideration in development of membrane for electrochemical device applications such as fuel cells batteries and water electrolysis. The role of membrane in electrochemical devices is crucial compared to the all other processes [19,20]. In the case of reverse osmosis, ultrafiltration, microfiltration, or nanofiltration the role of membrane is to act as a molecular sieve, whereas in case of electrochemical devices in spite of acting as a molecular sieve, membrane performs certain other key roles like it separates anode and cathode, prevents mixing of fuel and oxidant and also provides a conductive pathway. In case of electric field gradient, the membranes have to face some unusual challenges [21,22]. A schematic representation of membrane separation processes is represented by following Figure 1.

The usefulness of a membrane in a mass separation process is determined by its chemical, mechanical, thermal stability, its selectivity, and by its overall mass transportation rate. The chemical nature of the membrane's material is of prime importance when components with more or less identical molecular proportions and similar chemical or electrical properties that have to be separated. The mechanical, chemical and thermal stability of the composite membrane determines to a large extent, its useful lifetime especially when the feed solutions are of extremely low or high pH values or when frequent cleaning procedures of the membrane are required or when the process has to be carried out at elevated temperatures [23,24]. The mechanical properties of a membrane are of a special significance in pressure driven processes such as ultrafiltration and reverse osmosis. Ideally, a membrane should not change its properties when it is dried out or when the composition of feed solution has changed significantly. To significantly expand the use of membranes in mass separation processes beyond their present applications the membranes with more specific transport properties, longer lifetimes and higher flux rates are required [25].

3. CLASSIFICATION OF MEMBRANES

The membranes may be natural or synthetic, thick or thin, neutral or charged**.** Synthetic membranes can be subdivided into some categories such as organic (polymeric or liquid), inorganic (ceramic or metallic), and composites (organic-inorganic). The synthetic membranes are usually used for separation purposes in laboratories as well as in industries. In case of solid synthetic membranes, there are two types of membrane structures such as symmetric or isotropic and asymmetric or anisotropic. These membranes are classified, based on their surface chemistry, bulk structure, morphology, and processing method. The chemical and physical properties of synthetic membranes and a choice of driving force define a functional membrane separation process, and it utilized in separation process may be of different geometry and the respective flow configuration [26]. Synthetic membrane is fabricated by organic or inorganic materials, including solids such as metal or ceramic, homogeneous films (polymers), heterogeneous solids (polymeric mixes and mixed glasses), and liquids. Homogenous membranes are coherent ion-exchanger gels in the shape of disks, ribbons, etc. Their structure is that of an ion exchange resin and because of their uniform structure these membranes have been preferred for industrial approaches, while heterogeneous membranes consist of

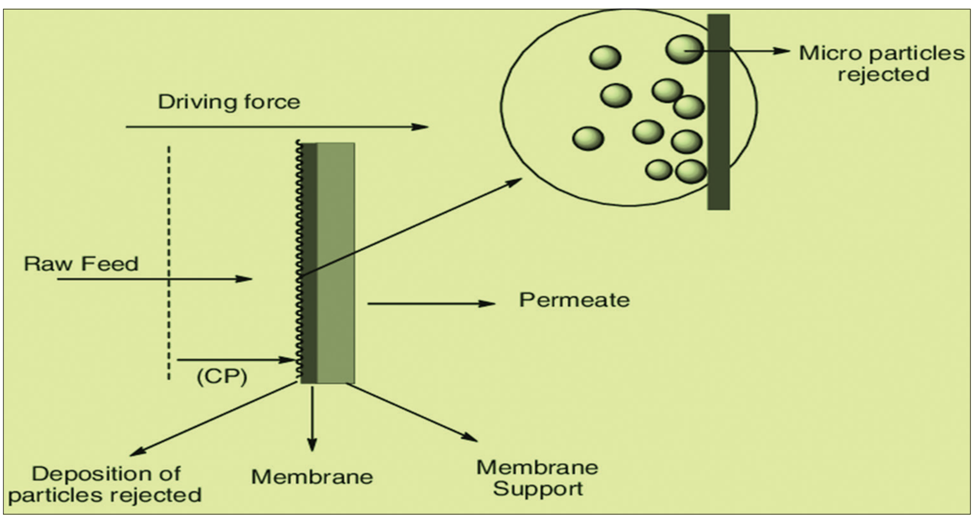

Figure 1: Schematic representation of composite membrane separation processes.

colloidal ion-exchanger particles embedded in an inert binder. As a result, only positively charged ions can migrate through cation exchange membranes, while anion exchange membranes allow the transportation of negatively charged ions. In homogenous membrane, it is an important feature to notice that the charges of membranes must be homogeneously distributed throughout the membrane body. The cation selective membranes are usually made by the cross linked attachment of polymer like polystyrene with metallic phosphate or tungstate, which should make a uniform distribution of composites. The anion selective membranes are usually made by mixing the polystyrene and metal containing quaternary ammonium ($NR^{+}_{4,}$) groups. Such type of anion exchange material can be synthesized, namely, the post-treating of polystyrene with mono chloromethyl ether and aluminum chloride which was used to introduce the chloromethyl group into benzene ring followed by the formation of quaternary ammonium by trirnethylamine [27,28].

In heterogeneous membranes, the finely powdered ion-exchange particles are not uniformly distributed throughout the membrane matrix. A stable heterogeneous membrane always show much finer unequal dispersion of ion-exchange particles that can be synthesized by mixing the polymer plastisol as well as ion exchanger inorganic compounds. On the basis of particles distribution and porosity, membranes are classified as polymeric membranes, ceramic membranes, liquid membranes, microfiltration membranes, ultrafiltration membranes, nanofiltration membranes, reverse osmosis membranes, polymeric-inorganic composite membranes, and ion-exchange membranes [29]. The following Figure 2 shows the schematic representation of membrane classification.

4. POLYMERIC-INORGANIC COMPOSITE MEMBRANES

Polymeric-inorganic composite materials have emerged as the most applicable engineered materials. These composite membranes are synthesized by the mixing of organic polymers with inorganic materials in a definite ratio of material percentages. Therefore, these membranes show the properties of both the used polymeric and inorganic materials. Polymer provides the flexibility and binding characteristics while the inorganic material gives the thermal stability and ion-exchange property. Mostly polymers such as polyvinyl chloride, polystyrene, and polyaniline are used that shows the stability and flexibility for the ideal membrane formation. Therefore, such composite membranes have indicated much application in various fields of chemistry, pharmacy, and material engineering. In most of the industrial processes such type solid polymeric and inorganic ion-exchange composite membrane are used for the process of purification, separation, and decontamination of aqueous and other ion containing solutions. Typical ion exchangers used to make the ion exchange membranes, namely, ion-exchange resins, zeolites, montmorillonite, clay, and soil humus [30]. Polymeric-inorganic composite ion-exchange membranes are widely used for fuel cell storage batteries, electrochemical separation, electro dialysis, electro deionization, etc., have drawn the attention of researchers in making ion selective membrane using organic-inorganic composite ion exchange materials due to their excellent performance for cation-exchangers. Cation exchange membranes have been widely explored by researchers over the past few years due to their commercial application's while anion exchangers are poorly reported and need proper investigations because of their importance in the field of environmental science for separation, identification, and determination of toxic anions from industrial waste and drinking water. There are also amphoteric exchangers which are able to exchange both the cations and anions simultaneously. These ion exchangers may be selective or nonselective which are used to bind certain ions or classes of ions depending on their physical and chemical properties [31,32].

5. APPLICATION OF POLYMERIC-INORGANIC COMPOSITE MEMBRANES

Membranes based technologies witnessed to a great extent of R&D efforts in recent years and have much potential to impact multimillion dollar industries in various sectors. Mostly they are used in food/drug production, separation of pollutants, water purification, textile, biotechnology, and to fuel cell markets. Polymeric-inorganic composite membrane constitutes a diverse collection of technological applications such as reverse osmosis, ultrafiltration, microfiltration, gas separation, electro dialysis, hemodialysis, and pervaporation [33]. However, in specific applications, the transfer of species across the membrane can also be a factor of consideration in the development of membrane for electrochemical devices such as fuel cells, batteries, and water electrolysis. The role of such membrane in electrochemical devices is crucial compared to all other filtration processes. In the cases of reverse osmosis, ultrafiltration, microfiltration, or nanofiltration the role of membrane is to act as a molecular sieve. Whereas in case of electrochemical devices in spite of acting as a molecular sieve the membrane performs certain other key roles like it separates anode and

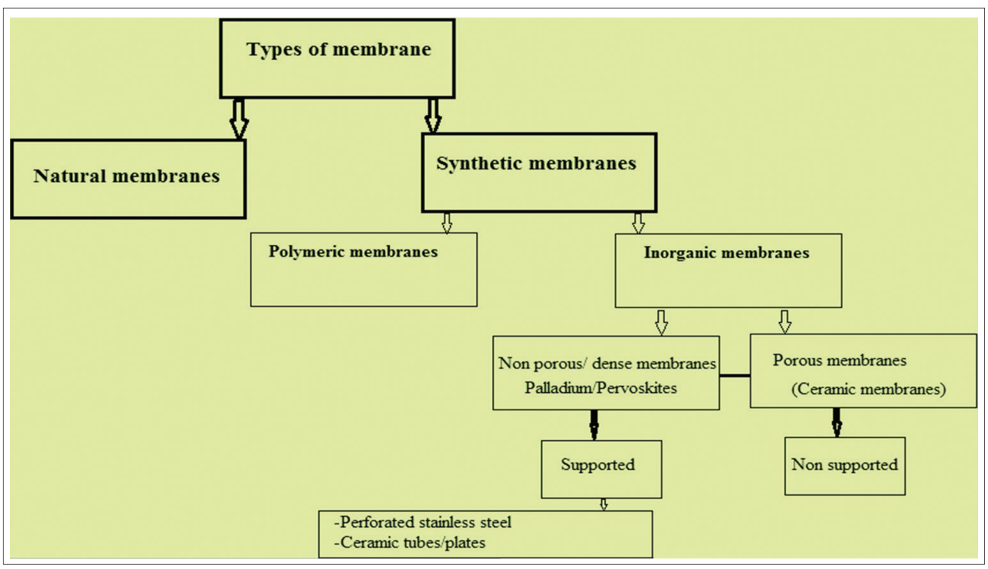

Figure 2: The schematic representation of membrane classification.

cathode, prevents the mixing of fuel and oxidant and it also provides a conductive pathway. In case of electric field gradient, the membranes have to face some unusual challenges [34,35].

Moreover, it finds applications in a wide variety of industrial, domestic, governmental, and laboratory operations. Such composite membranes have good ion-exchange capacity, higher stabilities, reproducibility, and selectivity for specific heavy metal ions, indicating their useful environmental applications. In general, these types of membranes have their applications in following disciplines

5.1. For Preparation of Qualitative Water

Membrane technology is widely accepted as a means of producing various qualities of water from surface water, well water, brackish water, and seawater. In the treatment of water for drinking purposes first of all pressure-driven membrane techniques are used. The choice of the suitable membrane process depends on the size of the removed contaminants and admixtures from the water. Desalination of seawater and brackish groundwater is often the way to obtaining drinking water. Significant improvements in technology and design of reverse osmosis, the availability of alternative energy sources, and the possibility of pretreatment and applied materials have caused the process to become environmental friendly source of fresh water in many regions of the world, particularly in those where the sources are limited. Nanofiltration and to some extent, the reverse osmosis (RO) is the methods of water softening, as well as to remove disinfection by products precursors and micro pollutants. To remove inorganic micro pollutants (nitrate, fluoride ions, boron, arsenic as well as chromium, and heavy metals), nanofiltration, reverse osmosis, electro dialysis and donnan dialysis, and ultrafiltration enhanced with polymers and surfactants as well as membrane bioreactors have been successfully applied. High pressure membrane processes (RO and NF) are an effective method for removal of dissolved organic compounds (DOC) in the treatment of natural waters. Scarcity of water, environmental requirements, and the simple logic of reusing water instead of discharging it are conditions, which call for increased the use of membrane technology in a multitude of applications [36].

5.2. Removal of Specific Constituents

Membrane technologies for removing particulate matter by micro and ultrafiltration, and dissolved substances by nanofiltration and RO are increasingly being used for wastewater reclamation in modern, urban water systems. Immersed micro and ultrafiltration membranes provide excellent pretreatment for RO, which can remove a wide range of dissolved constituents. For the reclamation of potable water, membrane technology must be followed by UV oxidation treatment for disinfection [37].

5.3. Nuclear Industry

Membrane processes are considered as potential methods useful in clean technologies that minimize the use of raw materials, rationalize energy consumption, and reduce waste production. They are capable to solve many environmental problems, among them problems related to nuclear technology field. Membrane processes have been already applied for liquid radioactive waste processing in many nuclear centers around the globe. Ceramic/composite membranes show a high chemical, temperature, and radiation resistance. Thermal process, namely, membrane distillation with the use of resistant porous membranes was proposed and tested for radioactive waste concentration. Other methods such as liquid membranes and electric processes with ion-exchange membranes as possible applications in nuclear industry are under development. Membrane methods were considered as alternative solutions for reclamation of different materials that can be recycled and reused [38].

5.4. Pulp and Paper Industry

Membrane filtration is today an industrial process in the pulp and paper industry. The rising demand for environmental protection, energy savings, and recovery of valuable products has accelerated the application of membrane filtration. These membranes are now commonly used in several areas of paper and pulp industry such as for the treatment of freshwater, chemical recovery, and treatment of bleaching effluents. Such membrane technology can also be used to complement or improve water recycling and closing the loop in the pulp and paper industry. Applications of membranes have been arranged under three headlines, effluent treatment, concentration, and fractionation. They have been adopted with the aim of decreasing water consumption, recovering and recycling valuable compounds, and reducing the environmental impact of pulp and paper mills [39].

5.5. Food Industry

The use of membrane technology as a processing and separation method in food industry is gaining wide application. Membrane separations can be used novel technology for processing new ingredients and foods. Membrane separations are considered green technologies. The major advantages of the technology are very little change in nutrition quality, the energy used in RO or ultrafiltration membrane is efficiently utilized as there is no phase change and the concentration of the food solution is established by applying high pressure to reverse the direction of flow. This technique is used to separate the low molecular weight solutes such as salts mono saccharides and aroma compounds from the food products. Some of the applications of this membrane technology are concentration and purification of fruit juices, enzymes, fermented liquors, and vegetable oils. Pressure driven membrane processes facilitate the separation of components with a large range of particle sizes. It is for this reason that they find wide range of applications in food processing industry [40].

5.6. Solvent Purification

Solvents are widely used in process industries such as chemical, petrochemical, pharmaceutical, food, biotechnology, and microelectronics. The recovery of spent solvents containing all types of impurities that are generating highly as well as marginally polluted solvent streams. Many current solvent purification and manufacturing practices utilize a traditional, energy intensive distillation based technology in the production or recovery of liquid chemicals. This method of processing is costly, inefficient and produces a significant volume of hazardous waste. Hence, it is replaced with membrane separation techniques working at room temperatures and able to achieve separations at the molecular level. Such chemical process intensification in solvent recovery and purification is expected to lead the significant improvements in product cost, materials usage, energy and waste reduction, as well as a decrease in risk and hazards [41].

5.7. Reagent Purification

Reagents such as water, ammonia, hydrochloric acid, nitric acid, perchloric acid, and sulfuric acid can be purified through membrane distillation process. Membrane distillation is a separation process where a micro porous hydrophobic membrane separates two aqueous solutions at different temperatures. The hydrophobicity of the membrane prevents mass transfer of the liquid, whereby a gas liquid interface is created [42]. Because of the difficulty in regenerating the starting material, relatively few derivatives of organic substances can also used by membrane purification. Membrane distillation can be used for the separation/concentration of non-volatile components, such as ions, acids, colloids, and macro molecules from aqueous flows, for the removal of traces of volatile organic compounds such as benzene, chloroform, and tri chloroethylene from water, and for the extraction of other organic components such as alcohols from diluted aqueous solutions.

5.8. Pharmaceutics and Medicine

Membrane technology is of major importance in medical applications. The membrane separation technologies of reverse osmosis, ultrafiltration, and microfiltration have been used to concentrate and purify both the small and large molecules. More recent applications of membrane technologies cover a broad range of separation, concentration and purification needs. For example, pharmaceutical waste streams can be treated by nanofiltration or pervaporation to detoxify them and reduce the volume of waste requiring incineration. Membranes are used in drug delivery, artificial organs, tissue regeneration, diagnostic devices, as coatings for medical devices, bio separations, etc. Membrane separation technologies are extensively used in downstream processes for bio pharmaceutical separation and purification operations through microfiltration and ultrafiltration processes. The new technique of membrane chromatography allows efficient purification of monoclonal antibodies. RO and nanofiltration techniques of membrane filtration are being combined with bioreactors and advanced oxidation processes to treat wastewater from pharmaceutical plants. Nanofiltration with organic solvent stable membranes can implement solvent exchange and catalyst recovery during organic solvent based drug synthesis of pharmaceutical compounds/intermediates. Membranes in the form of hollow fibers can be conveniently used to implement crystallization of pharmaceutical compounds in industries.

5.9. Soil Science and Technology

Regulatory compliance requires the cleanup of soils contaminated with toxic organic and metallic compounds. Soil washing with aqueous solutions transfers the contaminants from the solid matrix to the wash liquor, which then needs to be further treated. Membranes have not been used much for concentrating these wash liquors. However, several membrane techniques appear promising. In these techniques, the main objective is to find ways of concentrating the contaminants with much higher volume reduction. A combination of membrane processes, microfiltration and nanofiltration, was employed to enhance the removal of heavy metals from contaminated soil using acid leaching. Microfiltration was used to separate soil particles from the metal-containing leachate. The leachate was then processed using nanofiltration to reduce the leachate's volume and recover spent acid from the slurry. Results of the bench scale study demonstrated the advantages of incorporating membrane processes into soil treatment operations which should be faster and more complete removal of metals, reduced volume of waste products [43].

6. CONCLUSIONS

Membrane separation constitutes a diverse collection of applications involving technologies such as reverse osmosis, ultrafiltration, microfiltration, gas separation, electro dialysis, hemodialysis, pervaporation, and other business trends. The groundwater treatment is essential for the drinking purpose, which was found by the scenario of Indian groundwater quality. Desalination technologies create new sources of fresh water from sea or brackish water. This review summarizes the fundamental aspects and applications of RO and membrane distillation (MD) used for desalination. RO membranes are the leading technology for desalination of ground/Seas water because of their strong separation capabilities and exhibiting a great potential for treatment of waters worldwide. Polymeric-inorganic composite which comprises RO membrane used for desalination has been rapidly grown over the past 40 years to become the primary choice of new plant installation. In this review, the membrane has synthesized by die casting or metal casting (forcing molten metal under high pressure into a mold cavity) method, in which it is formed by mixing the organic polymers and inorganic particles together in definite ratio of percentages. Therefore, it is focusing that the synthesized membrane may classify into different groups due to their nature and application point of views which is clearly explained in the above paragraphs.

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***Bibliographical Sketch**

Dr. Mohd Arsalan has completed their M.Phil and Ph.D from Aligarh Muslim University Aligarh-India and he was an Assistant Professor in Department of Applied Chemistry Aligarh Muslim University Aligarh. He has nearly four years of teaching experiences as an Assistant Professor in the Department of Applied Chemistry, Dept of Chemistry and University Polytechnic, AMU, Aligarh. He has published more than 18 reputed research papers in the field of fabrication and characterization of composite membrane as well as synthesis of composite materials. His fields of research are Polymeric-Inorganic Composite Materials, Water Resources, Membrane Technology, and Wastewater Engineering. He has also visited some others countries for attending the International Conferences in the field of Desalination using Membrane Technology.