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High-throughput mixed-matrix membrane with superior anti-bacterial properties: A facile approach towards development of point-of-use water purification device

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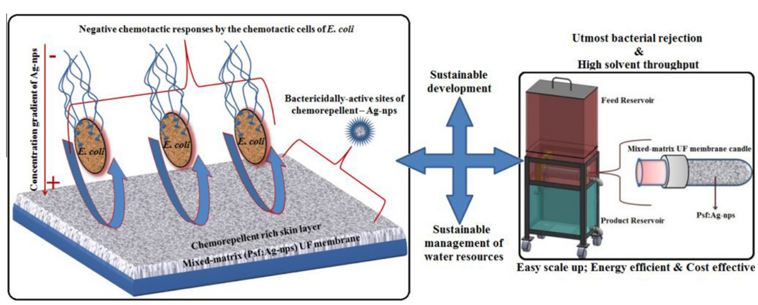
HIGHLIGHTS

- Mixed-matrix UF membrane as point-of-use bactericidal portable device developed.
- A methodology for energy-efficient and cost-effective domestic water purification developed.
- Bactericidal Ag-nps impregnated within a highly porous benign matrix of polymer.
- As a survival mechanism, bacteria experience negative chemotactic responses.
- Solvent throughput of 2500 LMH/bar and bacterial rejection of 99.99% achieved.

GRAPHICAL ABSTRACT



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ABSTRACT

A distinctive and impactful idea has been materialized to develop high-performance mixed-matrix ultrafiltration (UF) membranes using non-solvent induced phase inversion technique, utilizing various optimized compositions of polymer – polysulfone (Psf) and bactericidal nanostructured material, *i.e.*, silver nanoparticles (Ag-nps). Membranes in sheet-configuration with high void volumes were synthesized employing large amount of polyvinyl pyrrolidone (PVP) as porogen. The resultant porous morphology and surface chemistry, *i.e.*, surface hydrophilicity and electrokinetic features were assessed by instrumental techniques. The notable separation performances were observed as the membranes exhibited very high solvent throughput with reasonable anti-microbial activity. It has been substantiated that the nanoparticles upon impregnation within such a benign matrix of polymer exerted more pronounced mechanistic role towards anti-microbial efficacy on *Escherichia coli*, since as a survival mechanism the bacteria undergoing flagellar locomotory motion started experiencing negative chemotactic responses under the modified circumstances. With an objective of applying the salient features of such membranes, the facile methodical attempt was utilized in fabricating a domestic water purification device, with the membrane in candle-configuration for treatment of environmentally relevant aquatic media. It has been corroborated that our impactful approach towards development of an efficient water purification

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methodology (solvent throughput: 2500 LMH/bar and bacterial rejection: 99.99%) confirms the significance of membrane technology as a green and sustainable process with provision of easy scale up, energy and cost efficiency.

1. Introduction

Water resources are becoming increasingly scarce worldwide due to rising consumption of water at more than double the rate of the worlds' population growth, since the worlds' population is growing by about 80 million people per year with subsequent increase in the demand for fresh water by about 64 billion m^3 -per year, which will inflict nearly half of the global population to survive in high water stressed regions by 2030 [1]. In the coming decades, the possibility of having a ready access to safe drinking water may get boosted up by several other accelerating factors like rapid growth in urbanization [2] as well as industrialization, thereby increasing the level of contamination; over-exploitation of existing water resources, brought into by fast agricultural growth [3] coupled with financial and technological constraints and the alarming effects of global warming [4]. Different countries and regions with variant environmental, social and economic conditions have diverse needs with regard to the availability and quality of the vulnerable water resources, field conditions, access to technology and the types of technologies that suits appropriate in different circumstances. In accordance with the report of World Health Organization (WHO) and United Nations International Children's Fund (UNICEF), at present 1.1 billion people lack access to safe drinking water and about 2.5 billion people lack access to proper sanitation, nearly all of them in the developing countries [5]. The health burden of poor quality water seems enormous as it can be contaminated with a myriad of different toxic components, most particularly the major pathogenic organisms like viruses, bacteria and parasites which are responsible for life threatening water-borne diseases. These components have bio-cumulative, persistent and synergistic characteristics affecting health and function of the ecosystem as well as health and wellbeing of the humans. Leaky water distribution systems and deterioration of water quality upon aging (though water has been treated at the point of entry, POE) also account for such disastrous outcomes. Thus, a rising rivalry between water 'uses' and water 'users' for both existing fresh water as well as new water resources increases the importance of sustainable water management.

In the developing countries, most of the household water treatment technologies include chemical treatment methods like ozonation and chlorination, ultraviolet treatment, distillation etc. These current approaches have a large foot print and are not in a position to comply with the upcoming water quality standards of the developing urbanized and industrialized nations. In this context, membrane based separation processes have gained tremendous advancements in the recent past and continue to surge ahead as a frontier technology for sustainable water management. Furthermore, nanotechnology is identified as an area of science and technology that could play a potential role in addressing some of the inadequacies of the conventional membrane based separation processes [6]. The use of nanomaterials in several water treatment methods and devices could lead to cheaper, more durable and efficient membrane based water treatment technologies that can meet the needs of economically viable, reliable and selective separation systems [7]. However, it has been realized that a well-defined and well-engineered nanoscale material based water purification device for household applications should come into force to take care of safe water needs for all the sections of our society. Thus, the ease of utilization of functional properties of the inorganic nanomaterials as well as fabrication in porous matrices of various suitable organic polymers to achieve tunable physicochemical and superior macroscopical features turn the mixed-matrix membranes a promising contender for water treatment applications [8]. The familiar interplay between enhanced macroscopical features and possible formation of local defects in matrices of mixed-matrix membranes need to be balanced through optimization of several preparative conditions to derive the desired benefit of enhanced selectivity towards targeted species, without compromising with integrity of the membrane.

Numerous scientific investigations on membrane based separation processes have revealed that amongst different nanomaterials like Al_2O_3 [9], ZrO_2 [10], TiO_2 [11], SiO_2 [12], Ag [13–17] etc., Ag exerts an important role as a persuasive bactericidal agent in water disinfection. The anti-microbial efficacy of Ag, in zero-valent and ionic form, against bacteria, viruses and other eukaryotic microorganisms have been studied in great details [18]. The precise molecular mechanisms of action behind the bactericidal activity of Ag-nps against micro-organisms are not yet elucidated, but different inferences have been established in favor, through several interplaying factors *i.e.*, size, shape and physicochemical properties of the nanoparticles [19–21]. Several authors have provided different illustrations like dissolution of Ag^+ ions from Ag-nps, biological effects on formation of Reactive Oxygen Species (ROS) and extensive membrane damage etc [22,23]. The extremely large surface area and surface to volume ratio exponentially enhance the reactivity of the Ag-nps, which provides better and effective contact with micro-organisms, since the nanoparticles get attached to the cell membrane and subsequently penetrate inside the bacteria [24]. The Ag-nps of truncated triangular, spherical and rod shapes further exhibit varying anti-microbial efficacy on bacterial cells, as established through study of the inhibition of bacterial growth [20]. Numerous studies on Ag-nps embedded polymeric UF membranes have been reported in literatures, where, in all the attempts the mechanisms behind anti-microbial efficacy on bacteria have been attributed to aforementioned factors.

In an attempt made by Taurozzi et al. [14] the effects of variant casting mixture compositions resulting membranes with different porosities as well as the *ex-situ* and *in-situ* routes of nanoparticle incorporation were evaluated for Psf-Ag nanocomposite UF membranes. The study pointed towards extensive growth of bacteria in membranes having more porous features. In a study on nanocomposite UF membranes, made by Koseoglu-Imer et al. [25] it was noticed that there remains inherent trade-off between membranes' permeability and resistance towards growth of bacterial colonies as the former was significantly reduced to achieve a maximum of the later with progressive loading of Ag-nps. The effect of variation in sizes of Ag-nps on the performances and antibacteriality of Psf UF membranes was investigated by Mollahosseini et al. [26]. It was stated that the smaller size of the Ag-nps exhibited more efficiency in bacterial rejection. Basri et al. [27] reported the developments of antibacterial Polyethersulfone–Ag composite UF membranes, which were fabricated employing PVP of varying molecular weights as dispersants in the casting solution, where Ag^+ ($AgNO_3$) was reduced to Ag^o by PVP. It was quoted that a membrane prepared employing maximum loading of $AgNO_3$ and PVP of highest molecular weight exhibited maximum antibacterial activity by inhibiting the