



## Polysaccharide nanocomposites in wastewater treatment: A review

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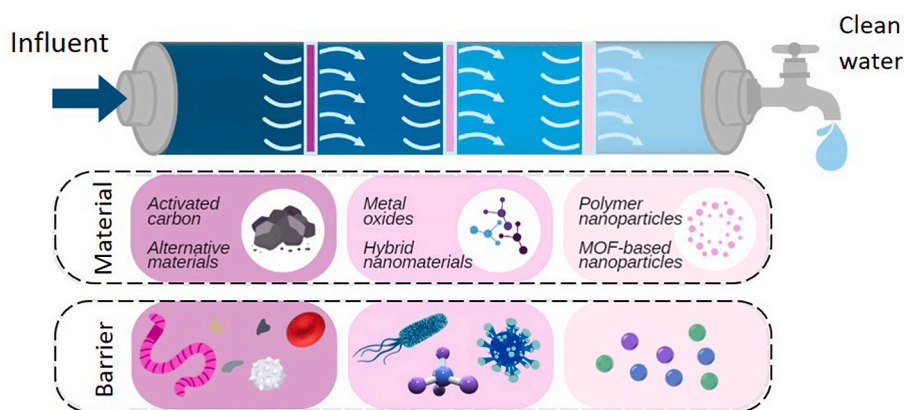
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### HIGHLIGHTS

- Polysaccharides (PSA) are eco-friendly biopolymers in wastewater treatment.
- Analyzed gas and liquid filtration, adsorption and proton exchange membranes.
- PSA nanocomposites aid in biological processes like antibiotic waste removal from water.
- Transitioning from lab to large-scale PSA use in wastewater treatment is pivotal.

### GRAPHICAL ABSTRACT



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## ARTICLE INFO

Handling editor: Sandeep K. Lakhera

## Keywords:

Polysaccharides  
Adsorption  
Membranes  
Nanocomposites  
Photocatalysis  
Wastewater treatment

## ABSTRACT

In modern times, wastewater treatment is vital due to increased water contamination arising from pollutants such as nutrients, pathogens, heavy metals, and pharmaceutical residues. Polysaccharides (PSAs) are natural, renewable, and non-toxic biopolymers used in wastewater treatment in the field of gas separation, liquid filtration, adsorption processes, pervaporation, and proton exchange membranes. Since addition of nanoparticles to PSAs improves their sustainability and strength, nanocomposite PSAs has gained significant attention for wastewater treatment in the past decade. This review presents a comprehensive analysis of PSA-based nanocomposites used for efficient wastewater treatment, focusing on adsorption, photocatalysis, and membrane-based methods. It also discusses potential future applications, challenges, and opportunities in adsorption, filtration, and photocatalysis. Recently, PSAs have shown promise as adsorbents in biological-based systems, effectively removing heavy metals that could hinder microbial activity. Cellulose-mediated adsorbents have successfully removed various pollutants from wastewater, including heavy metals, dyes, oil, organic solvents, pesticides, and pharmaceutical residues. Thus, PSA nanocomposites would support biological processes in wastewater treatment plants. A major concern is the discharge of antibiotic wastes from pharmaceutical industries, posing significant environmental and health risks. PSA-mediated bio-adsorbents, like clay polymeric nanocomposite hydrogel beads, efficiently remove antibiotics from wastewater, ensuring water quality and ecosystem balance. The successful use of PSA-mediated bio-adsorbents in wastewater treatment depends on ongoing research to optimize their application and evaluate their potential environmental impacts. Implementing these eco-friendly adsorbents on a large scale holds great promise in significantly reducing water pollution, safeguarding ecosystems, and protecting human health.

## Abbreviation list

<b>AC</b>	Activated Carbon	<b>Nd</b>	Neodymium
<b>Ag</b>	Silver	<b>NP</b>	Nanoparticle
<b>AOP</b>	Advanced Oxidation Processes	<b>PA</b>	Polyamide
<b>CB</b>	Conduction Band	<b>Pb</b>	Lead
<b>CD</b>	$\beta$ -cyclodextrin	<b>PES</b>	Polyether Sulfone
<b>Cd</b>	Cadmium	<b>PS</b>	Polystyrene
<b>CF</b>	Carboxymethylated fibers	<b>PMNC</b>	Polymer Matrix Nanocomposites
<b>CMNC</b>	Ceramic Matrix Nanocomposites	<b>PSA</b>	Polysaccharide
<b>CNC</b>	Cellulose Nanocomposite	<b>PVDF</b>	Polyvinylidene fluoride
<b>CoFe<sub>2</sub>O<sub>4</sub></b>	Cobalt Ferrite Oxide	<b>ROS</b>	Reactive oxygen species
<b>CS</b>	Chitosan	<b>SA</b>	Sodium Alginate
<b>Cu</b>	Copper	<b>Sc</b>	Scandium
<b>Fe<sub>3</sub>O<sub>4</sub></b>	Iron(II,III) oxide	<b>SiO<sub>2</sub></b>	Silicon oxide
<b>g-C<sub>3</sub>N<sub>4</sub></b>	Graphitic Carbon Nitride	<b>TiO<sub>2</sub></b>	Titanium Oxide
<b>GO</b>	Graphene Oxide	<b>Tm</b>	Thulium
<b>MB</b>	methylene blue	<b>Yb</b>	Ytterbium
<b>MgO</b>	Magnesium oxide	<b>ZIF</b>	Zeolitic Imidazole framework
<b>MMNC</b>	Metal Matrix Nanocomposites	<b>ZnO</b>	Zinc Oxide
		<b>ZSM-5</b>	Zeolite Socony Mobil-5

## 1. Introduction

The release of various pollutants on one front and the global pandemic on the other are posing a significant threat to the public health, prompting increased investments (Kordbacheh and Heidari, 2023; Maktabifard et al., 2023; Micah et al., 2023). As a result, governments and decision-making bodies are enacting stringent regulations to address these harmful agents (Watts et al., 2018; Al-Hazmi et al., 2023b; Lucero-Prisno et al., 2023). Correspondingly, wastewater treatment has become the matter of concern of researchers and technologists worldwide (Afify et al., 2023; Al-Hazmi et al., 2023b), such that a bewildering variety of approaches and methodologies such as coagulation (Lin et al., 2021), ion exchange (Peng and Guo, 2020), as well as membrane-based (Vatanpour et al., 2022b) or microorganism-based (Majumder et al., 2021) water purification, adsorption (Kegl et al., 2020) and photocatalysis (Nasrollahi et al., 2021; Salehian et al., 2022) have been developing and applying progressively. In addition to the need for the initial plant investments, these technologies are

resource-intensive to different degrees, and usually suffer from high operating expenses in terms of chemicals, energy and materials consumption (Holkar et al., 2016; Laureano-Anzaldo et al., 2021). When it comes with materials, plenty of nanoparticles and polymers (both neat and functionalized) have been in wastewater treatment plants and water purification processes for removal of diverse pollutants (Mahmoodi et al., 2017; Ismail et al., 2020; Taghizadeh et al., 2020; Naseem and Durrani, 2021). Furthermore, the use of single- and multi-stage processes could additionally support water purification and pollutant removal from the contaminated water sources (Lu et al., 2018; Al-Hazmi et al., 2021).

Polysaccharides (PSAs) are attractive bio-polymers best known for their functional groups such as hydroxyl (–OH), amide (–CONH<sub>2</sub>), sulfonate (–SO<sub>3</sub>), amine (–NH<sub>2</sub>), and carboxyl (–COOH) on the surface and/or in the main backbone (Abdelraof et al., 2020; Manzoor et al., 2022). Because of such a reactive nature, it could be possible to tailor-make PSAs by incorporation of a wide range of nanomaterials in order to develop nanocomposites for myriad wastewater treatment approaches (Alam and Christopher, 2018). Depending on homo- or

copolymer microstructure, PSAs can reveal a wide range of behavior and performance when interacting with living species – both flora and fauna, as well as microorganisms. Above and beyond, remarkable structural stability, biocompatibility and biodegradability can render the versatility of design to PSAs for biomedical and environmental applications (Seidi et al., 2021a; Vatanpour et al., 2022c). PSAs can also be grouped on the basis of their provenance – natural, chemically synthesized, or enzyme-aided semi-synthetic PSAs (Abdelraof et al., 2020). Their origin determines their composition, the type of functional groups, and the basic architecture of the PSAs. Algae, yield Carrageenan, alginate, chitin, chitosan and glycogen are of animal sources, while starch, galactomannans and cellulose are among vegetable-originated ones. The microorganisms are also from gellan and xanthan gum sources (Meng et al., 2018; Al Sharabati et al., 2021). Therefore, one can select them in the neat form or in combination with other counterparts to mitigate the harmful effects of pollutants, thereby controlling the pollution level and contamination of wastewater (Abdelhameed et al., 2022; Alkhadher et al., 2023b).

From the perspective of environmental science and technology, it is imperative for policymakers to pay significant attention to wastewater treatment (Al-Hazmi et al., 2023e; Kurniawan et al., 2023). This is especially crucial due to the rising levels of water contamination caused by a wide range of pollutants, including nutrients, pathogens, heavy metals, and pharmaceutical residues (Tavakoli et al., 2017; Liu et al., 2021a; Nasir et al., 2022). These pollutants endanger aquatic ecosystems and human health (Al-Hazmi et al., 2023c; Alkhadher et al., 2023a). In this sense, demand for developing eco-friendly methods enabling one to protect the environment has followed an ascending trend and become a challenging subject of research and development both locally and globally over the last two decades. One promising solution recommended and practiced by researchers was the use of eco-friendly PSA-mediated bio-adsorbents for wastewater treatment. PSAs, long sugar chains, are attractive adsorbents due to their biodegradability, non-toxicity, and abundant availability. They offer a sustainable alternative to harsh chemical treatments with harmful by-products (Fakhri et al., 2023). Wastewater contains various pollutants, including nitrogen, phosphorus, sulfate compounds, viruses, bacteria, heavy metals, thermal contaminants, radioactive substances, and pharmaceutical residues (Al-Hazmi et al., 2016; Ahmed et al., 2022). These pollutants can harm aquatic life, water quality, and the environment. Biological treatment methods serve microorganisms to break down organic matter, evidently effective in pollutant removal. Although conventional nitrification-denitrification and advanced anammox-based systems are widely used for nitrogen removal, they may not be versatile for all types of wastewaters (Al-Hazmi et al., 2022a). Therefore, application of natural and sustainable polymers like PSAs may bring about more possibilities for targeted water decontamination.

In a very recent work, we reviewed PSA-based membranes in view of materials, fabrication techniques, and separation performance (Vatanpour et al., 2022a). However, there are some disadvantages which need to be addressed and overcome, giving rise to PSA selection for efficient wastewater treatment plants. Some inferior characteristic, inter alia, include in hydrophilicity, low porosity, and poor mechanical properties (Zhu et al., 2019). Thus, there was a need for developing composites and nanocomposites based on PSAs to enhance the efficiency of wastewater treatment. Several nanocomposites based on or incorporated with PSAs have been used as surface modifier in wastewater treatment lines. Inorganic compounds such as metal oxides, structured carbons, as well as some biomolecules can be used as nanoparticles/nanofillers in developing different PSA composites and nanocomposites for a myriad applications (Pooresmaeil and Namazi, 2020; Oves et al., 2021). For instance, carrageenan membranes are used as aromatic encapsulation platform as well as sub-water superoleophobic membranes to remove organic dyes and metal ions (Perinelli et al., 2020; Prasannan et al., 2020). Furthermore, PSAs become long-lasting complex structures with higher mechanical strength in water when they are mixed with various

types of inorganic analogues. The functional groups on the PSA surface provide polymer composites with sufficient hydrophilicity, which is a sine qua non for wastewater treatment.

This paper attempts to emphasize the use of nanoparticle-aided PSA materials (or PSA nanocomposites) for water and wastewater treatment purposes. The incorporation of nanoparticles into PSAs can ameliorate the disadvantages of PSA like poor compatibility and affinity towards pollutants in aquatic environments, along with give other dimensions like antibacterial features to the PSA. Fig. 1 visualizes a broad image of progressive publications on PSA composites and nanocomposites for wastewater treatment. In this work, we first introduce PSAs and their nanocomposites on the basis of their structures, provenance, properties and synthesis routes. Then, wastewater treatment processes and earlier research in the field of PSA nanocomposites are discussed. Eventually, we keep eyes at the strengths, weaknesses, opportunities and challenges associated with the entrenchment of PSA nanocomposites in wastewater treatment to open new avenues of thoughts for future innovations and developments. We also deal with the practical aspects related to their cost-effectiveness, environmental sustainability, technological maturity and most importantly the pollutant removal efficiency. It can be seen that the term “chitosan” has been even more frequently used compared to “polysaccharide”, which evidences the importance of this biopolymer in water treatment surveys, or somehow pinpoints the lack of certainty of searching word “polysaccharide”, as the best keyword possible. In other words, chitosan has such a unique place among researchers that belongs to its own particular category in spite of taking a kind of place like other members of “polysaccharide” family.

## 2. Types of polysaccharides

PSAs are complex carbs formed by repeating sugar units (mono-saccharides) linked with glycosidic bonds (Hasanin, 2022). The diversity of reactive groups, high molecular weights and chemical composition makes them attractive as far as exploration of possibilities of harnessing them for different applications is concerned (Luo et al., 2021). They are vital macromolecules in plants, animals, and microorganisms, serving energy storage, structural support, and cell signaling. Common types include: 1) Starch - a major storage PSA in plants, consisting of glucose

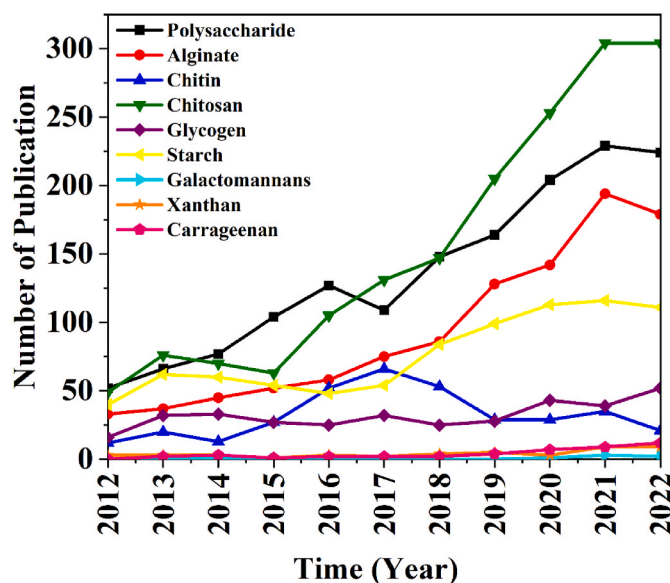


Fig. 1. The upward trend in publications directed at applications of PSA nanocomposites in general, and as per PSA type in wastewater treatment. These plots show an increasing interest in this field, especially for chitosan, alginate, and starch, during the past decade. However, the other PSA are not experiencing rapid increase of interest.

units in amylose (linear) and amylopectin (branched) forms; 2) Glycogen - a storage PSA in animals, similar to amylopectin but with more branching for rapid energy release; 3) Cellulose - a structural PSA in plant cell walls, made of linear glucose units providing rigidity; 4) Chitin - a structural PSA in arthropod exoskeletons and fungal cell walls, composed of linear *N*-acetylglucosamine units for support and protection; 5) Hemicellulose - a plant cell wall component with various monosaccharides like glucose, xylose, mannose, and galactose; 6) Pectin - found in plant cell walls, made of galacturonic acid units, involved in cell adhesion and gel formation in fruits; 7) Agarose - a seaweed-extracted PSA used in molecular biology for DNA fragment separation in gel electrophoresis; 8) Gums - PSAs from plants and microorganisms used in the food industry as thickeners, stabilizers, and emulsifiers. These examples illustrate the diversity of PSAs in the nature, each with unique structures and functions vital to living organisms.

Based on ionic charge, PSAs can be categorized as neutral (such as guar gum, starch, and dextran), positively-charged (like chitosan) and negatively-charged (like alginate and xanthan) (Ponnusami, 2021; Hassanzadeh-Afruzi et al., 2022b). The choice of suitable methods of preparation – precipitation, polyelectrolyte complexation, and self-assembly to name but three - are determined by their structural properties (Liu et al., 2008).

Cellulose, which is the most abundant biopolymer in the world, is a vital component of the walls of plant cells; and is also found in algae, oocytes and bacterial biofilms (Payne et al., 2015). A linear homopolymer composed of  $\beta$  (1  $\rightarrow$  4) linked repeating units of *D*-glucose, it is endowed with favorable properties, which make it amenable to various industrial applications (Hasanin, 2022). As mentioned earlier for PSAs in general, there are some challenges - hydrophilicity, water insolubility and crystallinity – which need to be taken on board and managed, if the favorable properties can be availed to the greatest extent (Liu et al., 2021b). This is all the more important when one considers the plethora of uses to which cellulose can be put - tissue engineering, diagnostic sensors, drug delivery, and hydrogels preparation (biomedical sector), paper products, food industry, and packaging (general industrial sector), electro-optical products, and energy storage (electronics and energy sectors), and yarn, fabric and clothing (textile sector) (Grishkewich et al., 2017; Mohammed et al., 2018; Sharma et al., 2019).

Edible and inexpensive starch, which comes at second place after cellulose, as far as natural abundance is concerned, has glucose in the form of beta-*D*-glucopyranose as its building block. Chitosan, which is derived from naturally-occurring chitin (found in the exoskeletons of arthropods – crabs, lobsters, squid, shrimps etc., insects and cell walls of fungi), is a cationic (positively-charged) PSA with an amino group. Its backbone includes randomly distributed  $\beta$  (1  $\rightarrow$  4) linked units of *N*-acetyl-glucosamine and *D*-glucosamine (Singh et al., 2017). Among its several favorable properties, to name can be the ability to chelate transition-metal ions, an excellent adsorption and film-forming capability, and bacteriostatic behavior innate, which is produced by brown algae (Phaeophyceae), and also some soil bacteria, consists of a linear chain structure of  $\alpha$ 1,4l guluronic acid and  $\beta$ 1,4d mannuronic acid; the two acids being found in different ratios (Hasnain et al., 2020). The physical properties of alginates strongly depend on their provenance (Lakouraj et al., 2014b). High biocompatibility, low toxicity, and facile gelation are attributed to the addition of divalent cations, like  $\text{Ca}^{2+}$ . They are widely used as film-forming compounds, due to a host of favorable properties, - eco-friendliness, biodegradability and inexpensiveness (Lakouraj et al., 2014a; Gheorghita Puscaselu et al., 2020), ability to thicken and remain stable in harsh environments, suspensions and gels (Raus et al., 2021; Thakur et al., 2018).

Xanthan, a heteropolysaccharide secreted by *Xanthomonas* bacteria, has a glucose backbone containing glucuronic acid ( $\text{C}_6\text{H}_{10}\text{O}_7$ ), mannose ( $\text{C}_6\text{H}_{12}\text{O}_6$ ), pyruvyl ( $\text{C}_6\text{H}_{11}\text{NO}_3$ ) and acetyl ( $\text{CH}_3\text{O}$ ) residues from a trisaccharide chain (Elella et al., 2021). It is water-soluble (Patel et al., 2020), has good dilution properties at low concentrations, and introduces a high degree of viscosity in aqueous media. Its rheological

properties remain unaffected by changes in pH values, ionic pressures and temperatures (Pettitt, 2020). Xanthan gum, sourced generally from corn, soya and wheat, on account of good solubility in both hot and cold water, stability conferred by hydrogen bonding, and pseudo-plasticity, is a material of choice for applications in industrial sectors ranging from food and pharma, to textiles to petroleum.

Gellan gum, an exopolysaccharide sourced from *Sphingomonas* elodea, is anionic (negatively-charged), water soluble (Zia et al., 2018), and has a very high molecular weight (approximately  $5 \times 10^5$  Da). It is essentially a series of tetra-saccharides of two  $\beta$ D-glucose residues - one from  $\beta$ D-glucuronic acid and the other from  $\alpha$ L-rhamnose. The original form of gellan gum is called a high acyl gelling agent and there are two acyl substitutes: acetate and glycerate. High acyl gellans are very flexible, resilient, indestructible and thermo-reversible. The removal of the acyl ( $\text{CH}_3\text{CO}-$ ) group results in the production of a low-acyl gellan gum, which has the ability to form hard, and stable gels (Palumbo et al., 2020).

Carrageenan, available in three forms of kappa, iota, and lambda, is an example of water-soluble heteropolysaccharide. It is well-accepted that the iota and kappa are thermo-reversible gels, while lambda behaves like highly viscous polymers. The Kappa clan of carrageenan family shows the minimum anionic nature among the three, which would be a decent choice for membrane applications (Dong et al., 2021). Locust bean and guar gum with sufficient quantities of galactomannans are progressively developing quickly as emulsion stabilizers and thickeners to fabricate edible membranes and coatings (Nwokocha, 2021). Glycogen is also best known as a highly-branched polymer apt for biological uses (Zhu et al., 2021). In the light of above explanations, one would have a wide spectrum of choice among PSAs for manufacturing nanocomposites for wastewater treatment in view of molecular and functional features.

### 3. Nanomaterials and polymeric nanocomposites

Due to advances in nanotechnology in the last few decades, natural polymers have been successfully modified into versatile and useful, structurally-improved nanomaterials (Sur et al., 2019). Vis-à-vis the traditional granular materials (milli-, and micro-levels), nanomaterials have superior mechanical/physical properties, which are topics of interest for nano-scientists (Khan et al., 2019). Bio-polymeric composites, in general, stand a notch above conventional filler-materials for example, when it comes to some key application-requirements like barrier properties and flame retardation (Jeevanandam et al., 2018; Ahmadi, 2019; Yan et al., 2019). PSAs, among bio-polymers, in addition to the afore-named advantages, confer additional benefits which have already been referred to earlier in the chapter (Ling et al., 2018; Zarintaj et al., 2023).

Polymer nanocomposites are typically a combination of nano-scale material with a macromolecule to guarantee both surface and bulk properties (Hasnain and Nayak, 2019). However, the manufacture of nanocomposites entails adequate process control to ensure that different materials which form the composite are compatible with each other at a molecular level, and in unison, acquire the end-properties (such as ductility and strength) desired for the applications the nanocomposites would be put to (Sun et al., 2020). However, once the processes and the control systems are in place, the outputs are, and will continue to be, in great demand in the manufacturing sector around the world (Abdelkhalek et al., 2022; Ibrahim et al., 2022).

With nanotechnology catching up, and nanocomposites likely to be in demand in the years to come, PSAs entering the fray augur well for a circular bio-economy in the future, when one considers the fact that the abiotic materials in vogue can be supplanted with biotic options with comparable or superior properties which include stability in harsh environments, and durability (Annu and Ahmed, 2018; Deepika et al., 2018; Alavi and Rai, 2020; Kanniah et al., 2022). Table S1 in the supplementary information (SI) provides a summary of different



PSA-nanoparticle combinations (the latter being used as reinforcements for the former), the associated preparation methods and applications.

#### 4. PSA nanocomposites in water and wastewater treatment

PSA nanocomposite particles – between 10 and 100 nm in size – can be blended with a polymer matrix. When distributed uniformly throughout polymers, the properties are influenced by their functional characteristics. The key parameters that play a crucial role in properties improvement include particle size, the type of biopolymer, the stoichiometric ratio of the components, the macromolecular backbone, and the variety of functional groups present (Ghanbari and Amanat, 2022). In the following sections, the utilization of PNCs in wastewater treatment processes – adsorption, photocatalytic treatment and membrane filtration/separation – are discussed (Koo, 2019).

##### 4.1. Adsorbents and adsorption

Adsorption is known as a process which can detach liquid or gas molecules from an environment or medium by collecting or trapping them physicochemically (Shah et al., 2017). This phenomenon in different ways based on the geometry of media, diffusion thermodynamics, and kinetics, and surface chemistry, which are thoroughly discussed elsewhere (Hato and Ray; Orasugh and Ray, 2022). Among possible ways is bulk diffusion, which is governed by dispersion and distribution fashion of adsorbent, besides interparticle diffusion which kinetically affects surface phenomena. The adsorption process thermodynamically seeks for an equilibrium state in between surface adsorption and desorption rates. Since the medium and the adsorbent have tendency towards equilibrium, but meanwhile are under the influence of impurities to be ‘adsorbed-away’, the phenomenon takes a complex and

less-known character (Eder et al., 2021). The adsorption equilibrium can be studied by taking recourse to an adsorption isotherm, adsorption isostere or an adsorption isobar. Furthermore, there are different types of adsorption isotherms, based on simplifying assumptions or complexities considered in theoretical and/or experimental investigations, which are studied well (Krese et al., 2018). Fig. 2 shows the overview of adsorption mechanism and adsorption process.

Carbon-based materials benefit from exceptional features such as selectivity that make them attractive in adsorption. Accordingly, for a large number of adsorption processes, a variety of carbon-based adsorbents have been used. Selectivity entails adsorbing certain categories of impurities like toxic heavy metals or organics for instance. This characteristic is also seen with PSA nanocomposite adsorbents (Seidi et al., 2021b), by virtue of its greater surface area and diverse functional groups, which improves the efficiency of adsorption or metal ions, dyes and dissolved gases (Fig. 3).

Magnetic nanoparticles (MNPs) are a promising class of adsorbents, best known for the capability of hosting and cleanup of impurities or pollutants followed by facile detachment from the environment applying external magnetic fields. Compared to the costly and repetitive utilization of non-magnetic nanoparticles as adsorbents, the use of MNPs for adsorption appears remarkable (Hassanzadeh-Afruzi et al., 2022a). Zhang et al. (2010) used octadecyl-functionalized ferrosferic oxide ( $\text{Fe}_3\text{O}_4$ ) MNPs coated with chitosan as adsorbent for cleanup of per-fluorinated wastewater, and reported higher dispersibility of MNPs in water because of chitosan shell-forming layer. Furthermore, MNPs enhance the adsorbent ability to block the penetration of naturally-derived macromolecules in polyhedral samples. Yadav et al. (2021) prepared a complex composite MNPs based on  $\text{Fe}_3\text{O}_4$  and carbohydrate polymers as surface modifiers, i.e., sodium alginate (SA) and  $\beta$ -cyclodextrin (CD)-modified activated charcoal (AC). The adsorption

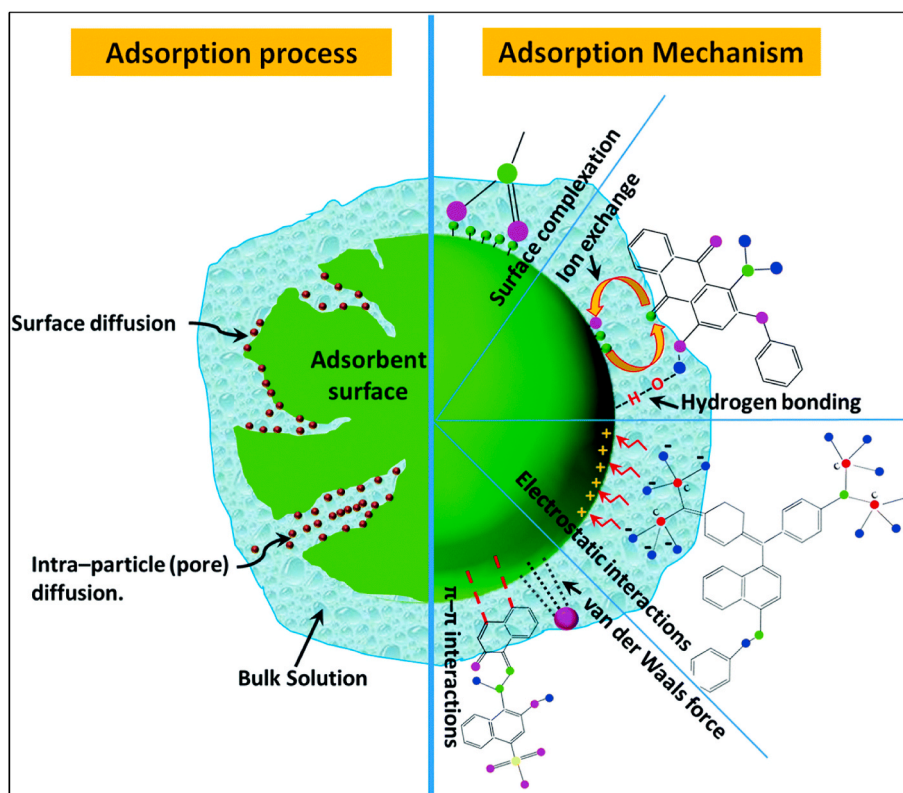
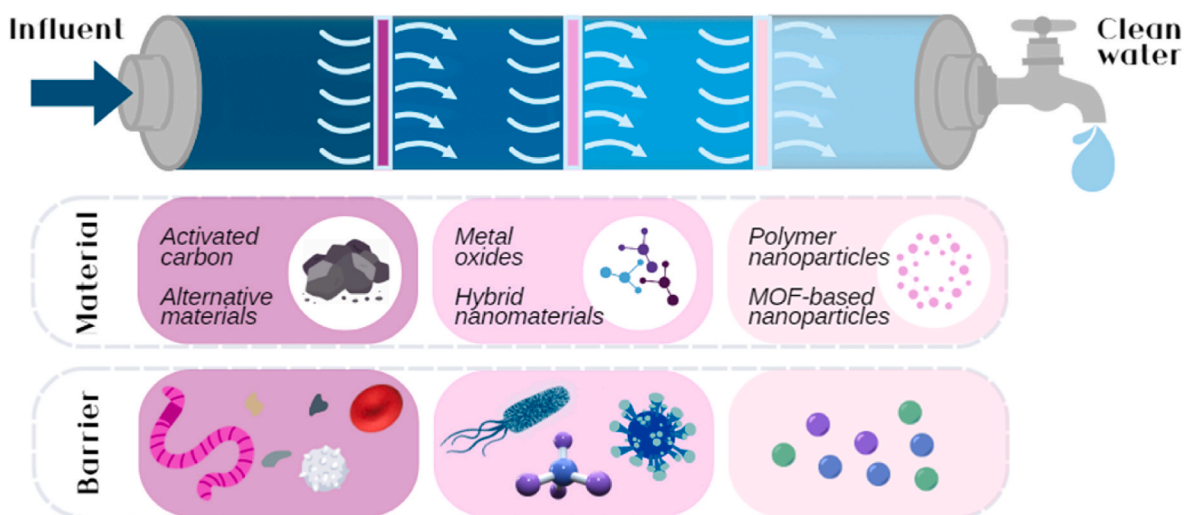


Fig. 2. Adsorption process and mechanism for PSA-based nano-adsorbents. Diffusion through bulk of solution, diffusion through porous structure of nanocomposite adsorbents, and surface diffusion constitute the adsorption process. In terms of mechanism, various physical and chemical interactions plays role in adsorption onto nanoscale PSA-based nanocomposites. These interactions include van der Waals interactions,  $\pi$ - $\pi$  stacking interaction, electrostatic forces, hydrogen bonding, and complexation interactions. Reprinted with appropriate permission from (Dutta et al., 2021). Copyright Royal Society of Chemistry.



**Fig. 3.** Overview of various nanomaterials used to make PSA-based nanocomposite adsorbents for separation of impurities by adsorption. Different types of nanostructures (such as carbon-based nanomaterials, MOFs, metal-oxide nanomaterials, and hybrid nanostructures) with various chemistries and different architectural features have been investigated as key constituents of PSA-based nanocomposite adsorbents.

efficiency of methylene blue (MB) as contaminant by the aforementioned of highly recyclable MNPs was above 99% over 90 min. Moreover, the reduction potential, and adoption capability for MB species was high leading to supreme separation performance. PNCs as hydrogel films have been shown to completely remove complex xenobiotics, metal ions and reactive textile dyes [95, 96]. Functional combination of cellulose and chitosan have been suggested as an effective alternative for removal of organic dyes from wastewater before discharge into the hydrosphere (Long et al., 2021). The separation was grounded on the effects of hydrophilic functional groups present on the backbone of the PSA, and cross-linkages between the branches and pollutants. Sharma et al. (2020) studied a PSA nanocomposite based on carboxymethyl cellulose/graphitic-carbon nitride/zinc oxide (CMC/g-C<sub>3</sub>N<sub>4</sub>/ZnO) by adopting the sol-gel technique for methyl violet adsorption from aqueous solutions. They reported that a adsorption as high as  $\approx 94.5 \text{ mg} \cdot \text{g}^{-1}$  can be achievable.

In another work, da Silva et al. (da Silva et al., 2020) showed the potential of polymer nanocomposite adsorbents based on a polyacrylamide surface modified with PSA for removal of an azo dyes. Likewise, Azhar et al. (Olad and Azhar, 2014) showed that the removal mechanism for reactive dyes using synthesized starch montmorillonite/polyaniline nanocomposites depends on the electrostatic attraction between the dye molecule and the nanocomposite. Elsewhere, (Vafakish and Wilson, 2019) chitosan-based tweezers were used to remove fluorescein and some more aromatic counterparts from wastewater solutions. They explained the successful process based on a proprietary categorization strategy that reinforces the adsorption features of chitosan through simple synthetic methods. Wang et al. (2019) employed several theoretical techniques, including molecular dynamics simulations and infrared spectroscopy. These methods were utilized to investigate the underlying physical interactions between okra PSA and methyl violet dye, which acts as a contaminant in an aqueous medium (Zare and Lakouraj, 2014; Hassanzadeh-Afruzi et al., 2023). The effect of the presence of several adsorbents in the medium, each having a different adsorption isotherm was discussed. Table S2 in the SM lists the foci of past research, of which the foregoing discussion is a small subset.

Metal-organic Frameworks (MOFs) are polymerized nanostructures, nowadays well-established in wastewater treatment besides other metal-oxides, which can remove a wide variety of contaminants (Salehi et al., 2023). Critical factors like the pH of the solution, the concentration of adsorbent in the medium, the initial concentration of the pollutants in the medium, and the temperature, as well as the electrostatic

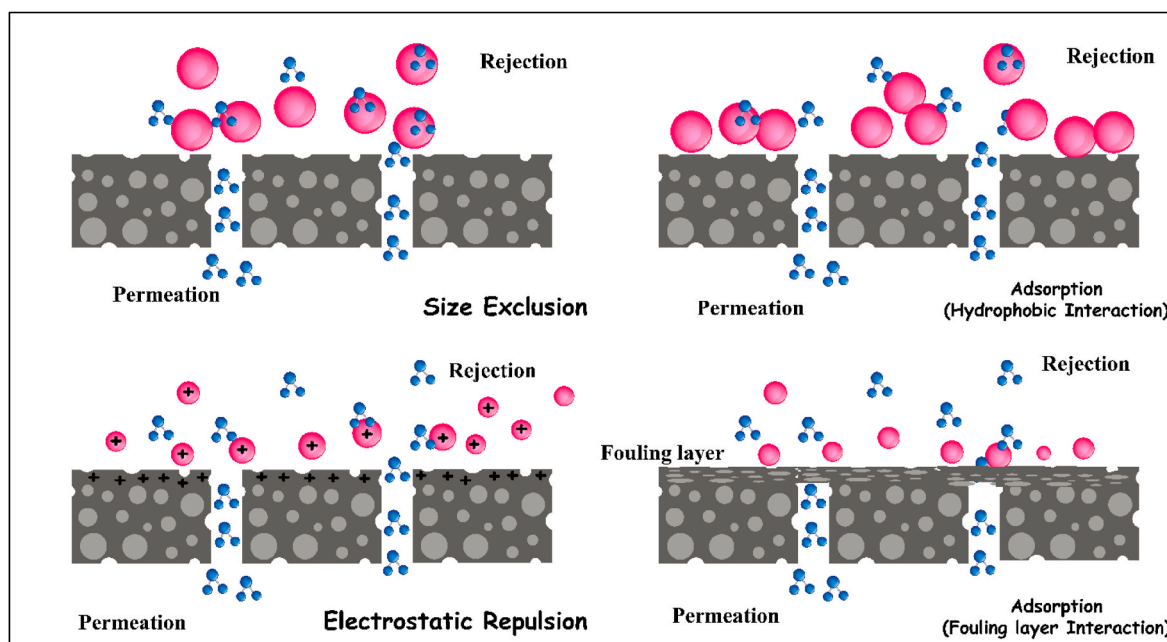
interactions, such as van der Waals forces, hydrogen bonding, and  $\pi$ - $\pi$  interactions, all govern the adsorption process.

#### 4.2. Membrane-based processes

Membrane technology has been, and is being, widely researched in both academic and industrial circles. Membranes facilitate a range of separation/filtration processes in the process sector, as well as the downstream environmental management operations (wastewater treatment, gas cleaning and soil remediation). A number of membrane-based technologies such as microfiltration, nanofiltration, reverse osmosis, and ultrafiltration have been widely used in separation operations (Vatanpour et al., 2020, 2021; Mahmodi et al., 2022; Sun et al., 2022). A detailed mechanistic explanation of the performance and purification efficiency of membranes in wastewater treatment is provided in Fig. 4. Contaminant removal is controlled by various parameters based on the properties of the membrane, the medium, and the pollutants to be handled, the operating conditions, and the tendency of the membrane to foul, among others.

The basic mechanism of membranes is to allow the solvent to permeate through its pores, while holding back the molecules which are larger in size than the pore-diameter (Ojajuni et al., 2015). However, studies have shown that other important physicochemical processes influence membrane filtration. Contaminant properties such as molecular weight and physical size (defined by length, width, diameter etc.), hydrophobicity or hydrophilicity, electrostatic charges on the particles, and their chemical structure are factors which need to be taken into consideration, on an equal footing with the properties of the membrane like pore size, surface charge measured by zeta potential, hydrophobicity or hydrophilicity as measured by the contact angle and surface morphology expressed as the roughness. Although these parameters are very well understood, further research is recommended to improve the degree of predictability of the performance.

Various polymers have been served in developing membranes for separation, filtration, and purification of wastewater, where chitosan and cellulose acetate are the most famous PSA ones. Similar to non-polysaccharide polymer membranes, difficulties have been associated with PSA-based membranes, such as biofouling, hydrophobic features, inappropriate removal efficiency, loss of mechanical properties upon aging, and loss of chemical stability can be highlighted. PSA nanocomposites are ideal membranes for wastewater treatment owing to their auspicious properties, including acceptable mechanical strength,



**Fig. 4.** Schematic illustration of various mechanism for membrane separation processes. In size exclusion mechanism molecules with dimensions lower than pore size are allowed to pass the membrane while bigger species are blocked. Membrane with surface electric charge accumulation repels species with the same surface charges while allowing the neutral species to pass through. The physical or chemical interactions between chemicals and membrane also affect the performance of membrane separation process. For example, for hydrophobic pollutants can interact with hydrophobic membrane surfaces through hydrophobic interactions resulting in adsorption and retention of these species on solid membrane. On the other hand, biofilm formation on the membrane increases the hydrophilicity of the surface, resulting the rejection of hydrophobic pollutant species.

appropriate chemical stability, facile film formation, appropriate selectivity and permeability, and also exceptional cleanup efficacy. The incorporation of metals and metal-oxide nanoparticles into a polymer matrix stabilizes its thermal and mechanical stabilities. In addition, salt removal efficiency and rate as well as fouling resistance enhance. As a manufacturing constraint, most of newly emerged membrane manufacturing strategies are not appropriate for scaling up to industrial applications.

Fig. 5 demonstrates method for preparation of films, such as electrospinning, phase inversion, and interfacial polymerization. [Abdelhameed et al. \(2021\)](#) obtained a highly porous adsorption membrane based on carboxylic acid (CA) and copper(II) benzene-1,3,5-tricarboxylate (Cu@BTC) MOF. Complexation between the functional groups of CA with  $\text{Cu}^{2+}$  metal centers in Cu@BTC MOFs endowed the final composite membrane with enhanced stability. Hydrogen bonds between carboxylic acid functional groups in ligands and hydroxy functionality of cellulose was responsible for stability of this structure. The porous membrane was fabricated using a two-step process. Initially, the CA membranes were synthesized, followed by immobilization of CuBTC on their surface using an ‘in-situ’ mechanism. To study the impact of MOF incorporation on membrane efficacy, the loading of CuBTC in matrix was varied from 20% to 60%. The obtained results showed that 40% incorporation of MOF results in the best efficacy. The structural properties of this membrane were studied using Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), and bacterial endotoxins test (BET). The obtained results proved effective immobilization of CuBTC onto the surface of CA membrane. SEM results showed pore size vary from 112.6 to 496.0 nm. According to the BET measurements, specific surface area for CA membrane increased significantly (from 78.4 to 965.8  $\text{m}^2 \text{g}^{-1}$ ) when incorporating 40% CuBTC. Accordingly, membrane’s adsorption capacity enhancement is closely related to the addition of CuBTC. Furthermore, it was found that the occurrence of aging during multiple cycles does not adversely affect the neither permeability nor selectivity of the membrane.

[Kuzminova et al. \(2021\)](#) developed a sustainable membrane from SA

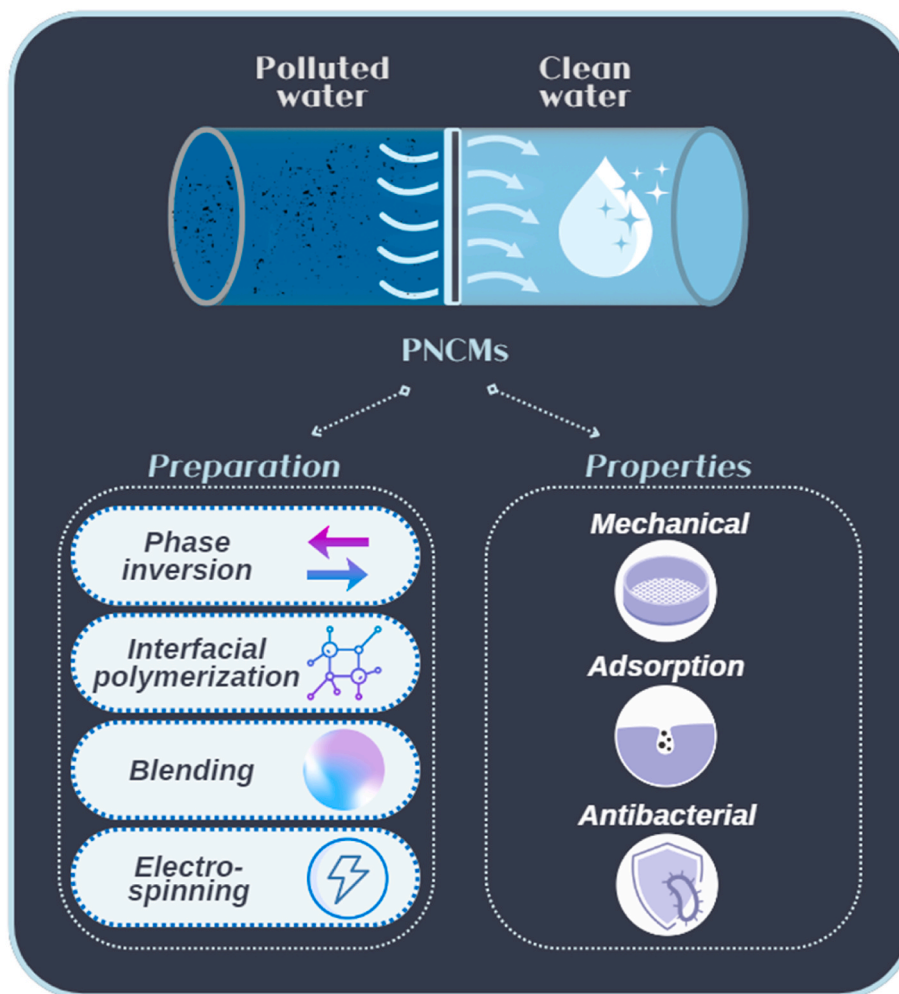
using FeBTC MOF to dehydrate isopropanol, generally-speaking for water removal. Incorporation of hydrophobic nanoporous FeBTC into the hydrophilic SA resulted in chemical stability in both aqueous and organic media. Thermogravimetric analysis (TGA) revealed that incorporation of 15 wt% of FeBTC into the SA results in an improved thermal stability. Additionally, it was found that the microstructural features of this nanocomposite significantly differ from those of the SA. Upon a flow rate of 174–1584  $\text{g m}^{-2} \cdot \text{h}^{-1}$ , the water content in the waste stream from the isopropanol dehydration process was around 99.99 wt%. Apart from their impressive separation efficiency, the majority of PSA/graphene oxide nanocomposites also demonstrate a remarkable ability to regenerate. This implies that they can maintain their efficiency over numerous cycles of operation, essentially allowing for multiple rounds of reuse ([Cui et al., 2023](#)).

[Lee et al. \(2013\)](#) used a PSA nanocomposite based on alginate, polyvinyl alcohol, and hematite ( $\text{Fe}_2\text{O}_3$ ) iron oxide to remove Cr(VI) from wastewater. The process reached equilibrium after 24 h with the highest adsorption capacity of 12.05  $\text{mg g}^{-1}$ . It was also revealed that the pH of the solution from which the hexavalent chromium was separated, influenced both the rate and the degree of adsorption; such that, a lower pH (acidic solutions) turned out to be more favorable to the separation process. Readers are referred to [Table S3](#) in the SI for related publications in which membrane-based processes are applied.

#### 4.3. Photocatalytic processes

Nanocomposites can generally be classified based on the matrix into metal matrix nanocomposites (MMNCs), ceramic matrix nanocomposites (CMNC), and polymer matrix nanocomposites (PMNC). A metal matrix (e.g., titanium [Ti], iron [Fe], copper [Cu], aluminum [Al], and nickel [Ni]) constitute the major part of MMNCs that can contain various soft or hard reinforcements such as oxides, nitrides and carbides, or sometimes a combination of such ingredients ([Gu et al., 2020](#); [Tariq et al., 2021](#); [Deng et al., 2023](#)). CMNCs based on nitrides and oxides are the first and metals are the second class of such materials ([Behera et al.,](#)





**Fig. 5.** Schematic illustration of the methods of PSA-based nanocomposite membranes preparation. Phase inversion include formation of thin nanocomposite film induced by selection of appropriate solvents. Polymerization of a monomer in interface between two immiscible solvents results in formation of a thin film in the interfacial polymerization strategy. Formation of nanocomposites membranes can be simply aided by solution blending of nanomaterials with PSA followed by evaporation of solvent (mostly water). In electrospinning process, a solution containing nanomaterials and PSA is pulled toward a plate upon application of an electric field. By adjusting electrospinning parameters, such as applied voltage, it is possible to finely tune the features (e.g., diameter) of electrospun nanocomposite nanofibers. This figure is designed based on the information represented in the article (Nasir et al., 2019).

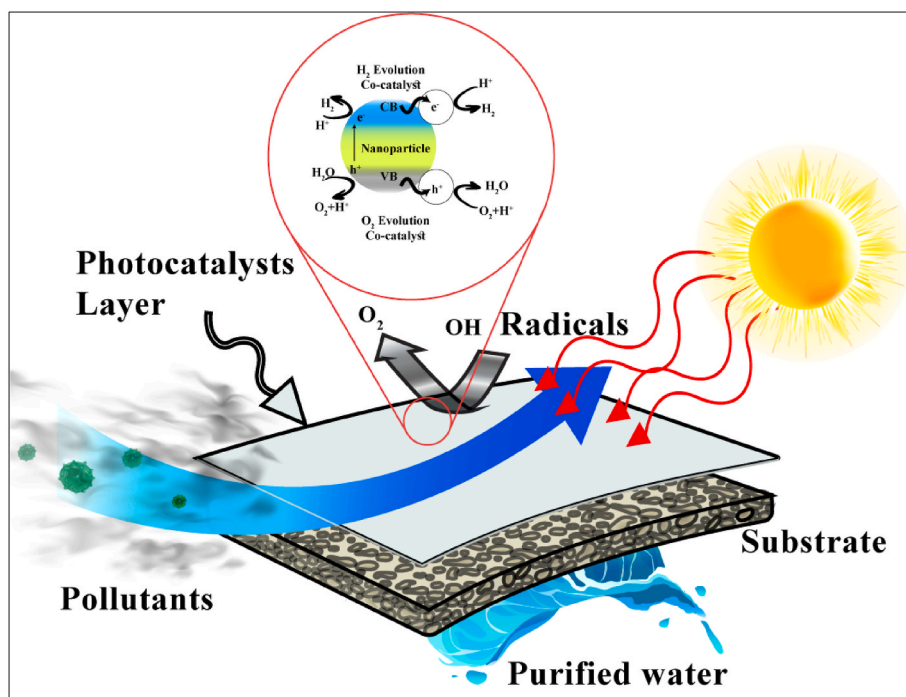
2020). On the other hand, in a PMNC a polymer serves as the matrix embedded with various reinforcement such as nanoparticles and nanofibers. Conventional wastewater treatment processes principally suffer from some shortcomings, mainly arising from limited efficiency. Accordingly, when it is required to remove contaminants with high efficiencies, it is essential to seek for novel strategies, which are not single-stage or classical. On the other hand, advanced oxidation water treatment technology can be categorized as irradiated (photocatalytic) or non-irradiated techniques (M'arimi et al., 2020). The latter comprises electrochemical oxidation, Fenton's reactions, ozonation and humid air oxidation. In photocatalysis, excitation induced by high energy photons results in formation of electron-hole pairs followed by creation of reactive radical species such as hydroxyl radicals (Simonsen et al., 2010). This leads to the complete mineralization of contaminants in the wastewater to non-toxic end products such as  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{NO}_3^-$  or  $\text{PO}_4^{3-}$  (Dey and Gogate, 2021).

PSA, or porous silica aerogel, can be regarded as a valuable component in photocatalytic advanced oxidation treatment systems. Thanks to its expansive surface area and robust adsorption capabilities, PSA can significantly reduce the generation of intermediates in the photocatalytic process (Deng et al., 2023). It also facilitates easy and rapid recovery and reuse of photocatalysts, with or without regeneration

(which can be looked upon as an economic and an environmental benefit; further, photocatalytic oxidation is clearly superior to traditional chemical oxidation from an environmental perspective). Heterogeneous photocatalysis can be carried out with natural sunlight – a promising contribution to green chemistry (Ma et al., 2018).

The integration of the PSA into the photocatalyst not only enhances the efficiency of photocatalysis but also mitigates the recombination of electron pairs generated either photochemically or electrochemically (Asadollahi et al., 2017; Guo et al., 2022). Taking a broader view, the integration of nanomaterials with polymeric substrates has led to advancements in numerous fields such as chemistry, heat resistance, optics, electronics, and resilience in challenging environmental conditions. Additionally, this fusion has bolstered the ability to produce thin films and bolstered mechanical strength, owing to the flexibility and robustness of polymeric materials (Hassani et al., 2017; Joseph et al., 2023; Soleimani-Gorgani et al., 2023). In order to obtain a homogeneous and well-dispersed polymer substrate with chemical, mechanical and thermal stability, the formation of an inorganic skeleton under mild operating conditions, and binding of the nanomaterial to the polymer matrix are prime requisites. In this method, the photocatalyst is separated from the reaction medium by simply removing the thin film from the aqueous medium, thus obviating the need for filtration (Su et al., 2021). While





**Fig. 6.** Mechanism of photocatalytic separation process for PSA nanocomposite nanoparticles. High energy photons of incident light are absorbed by photocatalytically active layer of PSA nanocomposite nanoparticles, resulting in creation of electron-hole pairs ( $h^+e^-$ ). Electrons released from conduction band (CB) promote reduction process like hydrogen evolution. In addition, holes formation in valence bands (VB) triggers oxidation process like oxygen evolution phenomenon. Reactive radicals, such as hydroxyl, can induce degradation of pollutants.

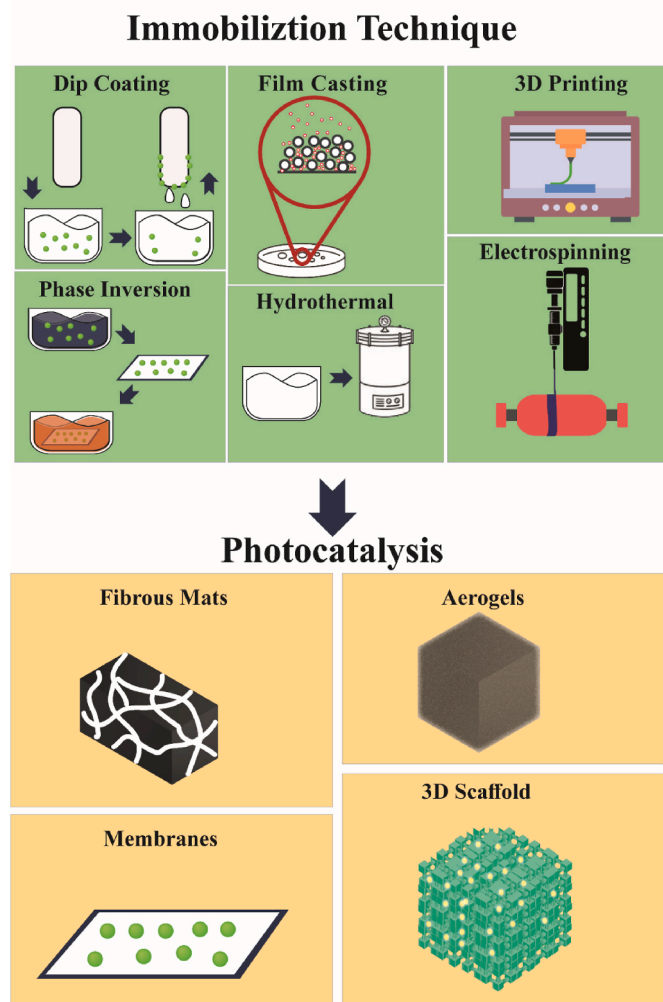
**Fig. 6** illustrates the mechanism of PNC-mediated photocatalysis, it would be apt to describe it briefly. Electron-hole pairs are formed due to the absorption of photons with the same or greater energy than the photocatalyst band-gap. The highly-oxidative holes produce radicals – hydroxyl ions for instance. Electrons meanwhile are repelled to positively-charged ‘acceptors’ having a higher reduction potential than the semiconductor conduction band.

While stability-of-structure has been emphasized in many publications (Ainali et al., 2021), very few have written about the admixtures of natural polymers in this regard. The technique used to immobilize nanoparticles on the support has a significant impact on the photocatalytic activity of nanoparticles and should thus be carefully chosen based on the utilized substrate and the pollutant/s to be removed from the wastewater (Hasanin et al., 2021; Nemiwal et al., 2021), and a handful of them have been dwelt upon – the sol-gel method (Khodadadi, 2016), which largely consists of film casting and dip-coating methods (Pinho et al., 2015; Farshchi et al., 2019) hydrothermal treatment (Li et al., 2020), plasma treatment (Park et al., 2011), electrospinning (Tu et al., 2019), electro-spraying (Korina et al., 2013), and 3D printing (Bergamonti et al., 2019). Different experimental techniques are used for the evaluation of the performance of the photocatalyst – for instance, the comparison of the performance of different architectures of chitosan-based materials including films, bilayers, 3D printed scaffolds, and fibers (Fig. 7). Reusability is also a desirable feature, and in many of the experiments with chitosan-based catalysts, most of them retained their effectiveness for 3–6 cycles, though in some cases, there was a marked deterioration after the second cycle. Several approaches – sol-gel, phase-inversion, and casting – were also used to develop cellulose-based frameworks for the immobilization of nanoparticles. Gelation techniques are frequently used to prepare alginate-based photocatalysts in a range of morphologies such as fibers, membranes, hydrogel spheres, and sheets (Subbiah and Palanisamy, 2022).

Lefatshe et al. (2017) fabricated a zinc oxide nanostructure in the cellulose host polymer through the casting method in the field to improve the antimicrobial and photocatalytic efficiency of ZnO. Organic

dyes are often degraded on the surface of a photocatalyst, making surface area an essential factor in photocatalysis. Their results show that MB photodegraded more easily in ZnO dispersed into the cellulose, than in the presence of pure zinc oxide. The apparent rate constants ( $K_{app}$ ) were derived in this paper as the slope ‘k’ of the linear regression equation [ $\ln(C_0/C) = kt$ ]; and were reported as 0.1013 and 0.1174  $h^{-1}$ , for ZnO and ZnO/CNC respectively. The higher value for the latter indicates that it has more active sites for photocatalysis vis-à-vis the former. The degradation follows a first-order kinetics, thanks to the porous three-dimensional structure and high surface area offered by cellulose, which augments the number of active sites available and thereby increases the reactivity. Still on the structure which plays a vital role in the photocatalytic process, Ma et al. (2021) demonstrated that three-dimensional g-C<sub>3</sub>N<sub>4</sub>@-cellulose aerogel functions effectively as a photocatalytic semi-conductor; with its bandgap (2.70 eV), conduction band (–1.3 eV) and valence band (1.4 eV) providing enough redox potential. More specifically, the results from that paper confirmed that a g-C<sub>3</sub>N<sub>4</sub>@-cellulose aerogel composition with a 30:70 ratio had outstanding photocatalytic activity for the breakdown of methyl orange and MB. Table S4 in the SI is a brief summary of findings from PSA nanocomposite-based photocatalytic experiments conducted in the past by researchers around the world.

While ensuring a high degree of performance by way of a good dispersion of the photocatalyst in the solution is a top priority, recycling the photocatalyst is also nonetheless important from economic and environmental perspectives. The first-named priority can be addressed by making sure that the active surface area is increased by utilizing suitable supports. This can also be achieved by transforming the photocatalysts into nano-fibers. Some supports have been reported to exhibit good hole-scavenging capabilities; and thus aid in the electron-hole separation during the photocatalysis (Sohouli et al., 2022).



**Fig. 7.** Schematic representation of various techniques that can be used for fabrication of PSA nanocomposite-based photocatalytic membranes. Various forms of photocatalytic membranes based on PNC. This figure is designed based on the information represented in the article (Jouyandeh et al., 2021).

### 5. Eco-friendly PSA-mediated bio-adsorbents for wastewater treatment

Generally, wastewater contains nutrients (i.e. nitrogen, phosphorus, and sulfate compounds) (Grubba et al., 2022; Al-Hazmi et al., 2023d), pathogens (i.e. viruses and bacteria) (Al-Hazmi et al., 2022b, 2023b) as well as, heavy metal, thermal, radioactive contaminants and pharmaceutical residues (Al-Hazmi et al., 2016; Wu et al., 2023). Biological treatment uses microorganisms to break down organic matter and solids in water. Common methods include activated sludge, trickling filters, and rotating biological contactors. Microorganisms convert organic matter into biomass and gases like carbon dioxide (Al-Hazmi et al., 2022c). These processes are vital for wastewater treatment to remove pollutants (Derwis et al., 2023). Two commonly used methods are conventional nitrification-denitrification and advanced anammox-based systems (Al-Hazmi et al., 2023c). Traditional nitrification-denitrification methods serve as a preventive measure against nitrogen-related problems such as eutrophication, which can be detrimental to aquatic ecosystems by causing oxygen depletion (Sallan et al., 2023). Anammox-based systems directly convert nitrogen compounds into nitrogen gas without oxygen, offering a promising approach to sustainable nitrogen removal in wastewater treatment and efficient water management (Al-Hazmi et al., 2021). Challenges include specific

conditions required and may not be suitable for low-strength wastewater with low ammonium content (Lu et al., 2018; Pereira et al., 2019). Nevertheless, anammox-based systems are innovative and offer a sustainable solution.

Ammonia, a prevalent nutrient pollutant in wastewater, damages aquatic ecosystems and their inhabitants. Elevated levels boost algae growth, reducing oxygen and endangering aquatic life. (Ong et al., 2022) investigated affordable green polymer (TEMPO-oxidized cellulose) for ammonia ( $\text{NH}_4^+$ ) removal in wastewater for fish farms. At 0.78 mmol/g carboxylate content, cellulose exhibited an adsorption capacity of 8 mg/g (or 9.5 mg/g per Langmuir isotherm) at pH = 7.

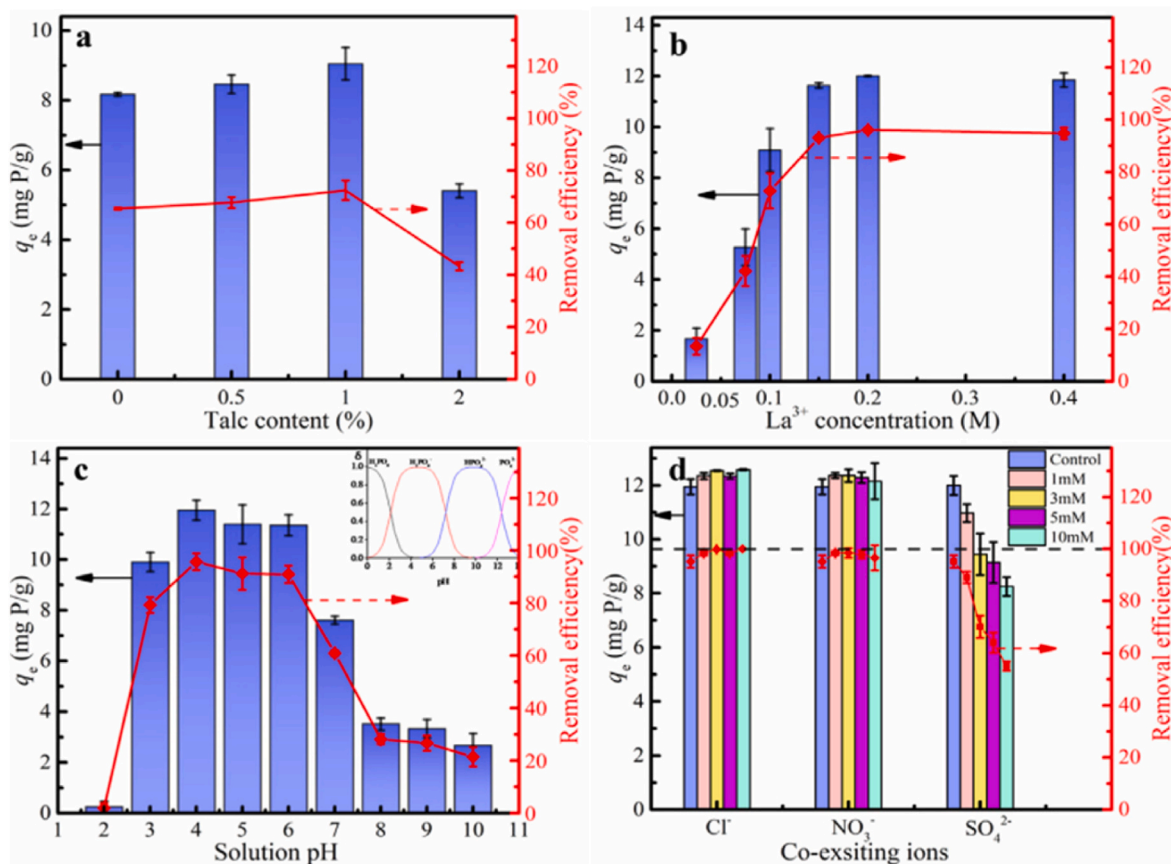
Phosphates are vital for aquatic ecosystems, supporting plant and algae growth at the base of the food chain. However, human activities release excessive phosphates, leading to eutrophication (Kock et al., 2023). To protect aquatic ecosystems, it must reduce phosphate input through better agriculture, improved wastewater treatment, and phosphate-free cleaning products. One example of a natural PSA with phosphate removal capabilities is chitosan, derived from the shells of crustaceans like shrimp and crabs (Balakrishnan et al., 2023; Fakhri et al., 2023) reported that chitosan-based materials have been used in water treatment processes as an effective method to remove phosphates. When chitosan is added to water or wastewater, it can bind to the phosphate ions, forming insoluble complexes that can be easily separated from the water through sedimentation or filtration. Chitosan/lanthanum hydrogel beads with glutaraldehyde as the cross-linking agent offer affordable phosphorus removal (Koh et al., 2022). They demonstrated maximum adsorption of 107.7 mg/g at pH 4 with a 6-h contact time. These beads effectively removed toxic lead, zinc, and copper from water through ion exchange and electrostatic attraction. Even after the fifth cycle, >80% phosphorus removal was maintained.

Wang and coworkers (Wang et al., 2020) investigated the influence of different talc encapsulated salts alginate (TAL) content (0%–2%) on phosphate removal using TAL beads is shown in Fig. 8a. Increasing talc content from 0% to 1% led to improved phosphate removal by TAL beads. Notably, TAL-2 and TAL-0 removed around 72% and 65% of the initial phosphate, respectively, with this enhancement attributed to the combined influence of  $\text{La}^{3+}$  and talc. The study investigated how different  $\text{La}^{3+}$  concentrations, ranging from 0.025 M to 0.04 M, affected phosphate uptake, as shown in Fig. 8b. Increasing  $\text{La}^{3+}$  concentration from 0.025 M (TAL-4) to 0.15 M (TAL-7) significantly boosted TAL bead performance. The adsorption capacity increased from 1.69 to 11.59 mg P/g, and removal efficiency improved from 13% to 93%. This highlights the dominant role of  $\text{La}^{3+}$  in the phosphate adsorption process.

In general, the pH sensitivity of the adsorbent can be attributed to various factors, including phosphate species presence, TAL-7 bead surface charge, and coexisting ions' influence. Specifically, at pH 4, phosphate primarily exists as  $\text{H}_2\text{PO}_4^-$  (Fig. 8c). As the solution's pH decreases from 4 to 2,  $\text{H}_2\text{PO}_4^-$  gradually converts into  $\text{H}_3\text{PO}_4$ . This conversion hampers phosphate adsorption by reducing the electrostatic attraction between phosphate ions and  $\text{La}^{3+}$ , resulting in poor adsorption performance. Additionally, in the pH range of 7–10,  $\text{HPO}_4^{2-}$  becomes the dominant phosphate species.

Fig. 8d illustrates the impact of co-existing ions on TAL-7 bead phosphate uptake. Typically, aqueous anions are known to hinder phosphate adsorption by competing for active sites. Surprisingly, Cl and nitrate ( $\text{NO}_3^-$ ) were found to enhance phosphate uptake by TAL-7 beads. When these anions penetrated the positively charged hydrogel, they neutralized it, resulting in a more porous and open matrix with increased channels and larger pores. This structural modification, in turn, promoted the adsorption process.

Adsorption mechanism based on the results analyzed above, (Wang et al., 2020) indicated that  $\text{La}^{3+}$  activity sites likely form via protonation, as shown in Fig. 9, ( $\text{TAL-La-OH} + \text{H}^+ \rightarrow \text{TAL-La-OH}^{2+}$ ). In their study, it is proposed that  $-\text{OH}^{2+}$  replacement at metal binding sites happens more easily than  $-\text{OH}$  replacement at low pH, enhancing ligand exchange. Thus,  $\text{La-O-P}$  inner-sphere complexes are likely generated via



**Fig. 8.** The impact of various factors on the adsorption performance of talk encapsulated salts alginate (TAL) beads, specifically focusing on: (a) talc content, (b)  $La^{3+}$  concentration, (c) solution pH, and (d) co-existing ions. These experiments were conducted with consistent parameters, including an adsorbent dosage of 0.4 g/L, a pH level of 4, an initial concentration ( $C_0$ ) of 5 mg P/L, a contact time ( $t$ ) of 24 h, and a temperature ( $T$ ) of 25 °C. Reprinted (reproduced) with permission from (Wang et al., 2020), Elsevier.

ligand exchange, supported by XPS analysis. In contrast, Talc, especially its Mg–O band, aids phosphate uptake in metal/oxide/phosphate complex formation. Energy dispersive spectrometer analysis reveals ion changes, including Cl, during adsorption. As pH rises from 8 to 10, La active sites may shift toward forming La–O coordination bonds with phosphate oxygen ions, especially under alkaline conditions (Fig. 9).

Essential trace elements are required by organisms in small amounts for normal physiological functions. They are typically incorporated into metalloproteins and enzymes, where they serve as cofactors and activators, playing crucial roles in catalytic reactions. Iron ( $Fe^{2+}$  and  $Fe^{3+}$ ) and manganese ( $Mn^{2+}$ ) are examples of essential trace metals that participate in enzyme activation and redox reactions (Bogacki and Al-Hazmi, 2017; Xu et al., 2022). On the other hand, heavy metals like cadmium ( $Cd^{2+}$ ), mercury ( $Hg^{2+}$ ), and lead ( $Pb^{4+}$ ) are known for their toxicity and harmful effects on living organisms (Jaishankar et al., 2014). They are not considered essential and can be detrimental to enzymatic activities and cellular processes. In fact, exposure to high levels of these toxic heavy metals can lead to various health issues and environmental pollution.

It is essential to distinguish between essential trace elements that are beneficial at appropriate levels and toxic heavy metals that should be avoided or carefully regulated due to their potentially harmful effects. Mixing essential trace elements with toxic heavy metals can lead to a misinterpretation of their role in biological processes and could have serious consequences (Feng et al., 2017). Elevated concentrations of heavy metals impede the functionality of anammox processes, leading to a direct reduction in the efficiency of nitrogen removal within anammox-based systems (Al-Hazmi et al., 2023a; Derwis et al., 2023; Khanzada et al., 2023).

In contrast, (Pagliaccia et al., 2022) found that extracellular polymeric substances in anammox granular can adsorb and remove heavy metals like  $Pb^{4+}$ ,  $Co^{2+}$ ,  $Ni^{2+}$ , and  $Zn^{2+}$  from waste sludge, enhancing the efficiency of anammox-based systems for wastewater treatment. This presents an added advantage in achieving economic and environmental sustainability in wastewater treatment (Zare et al., 2018). However, PSA-mediated bio-adsorbents may also aid in reducing heavy metal concentrations to safe levels, promoting proper microorganism function in wastewater.

(Fakhri et al., 2023) indicated that PSA nanocomposites can positively affect biological processes in wastewater treatment plants by i) Increasing adsorption capacity for contaminants (heavy metals, organic dyes, compounds) due to high surface area and functional groups; ii) Supporting biofilm formation, enhancing microbial activity, and removing organic matter in biological treatment; iii) Acting as a carbon source, aiding nutrient removal (nitrogen and phosphorus); iv) Exhibiting antibacterial properties, reducing harmful bacteria in wastewater and improving microbial dynamics; v) Assisting in floc formation, leading to better removal of suspended particles and clearer treated water; vi) Reducing membrane fouling in membrane-based treatment, improving filtration efficiency and longevity.

Cellulose-mediated adsorbents show impressive water pollutant removal abilities. Cellulose, Earth's primary PSA, makes up ~50% of plant biomass, forming plant cell walls. Its exceptional quality features, such as biodegradability, strength, and low toxicity, has led to its widespread use in various industries. (Etale et al., 2023) indicated that a biodegradable cellulose such as carbohydrate polymer ( $C_6H_{10}O_5$ )<sub>n</sub> with glucose monomers linked by  $\beta$ -1,4-glycosidic bonds, is essential in plant and algae cell walls. Its strength, non-toxicity, and exceptional



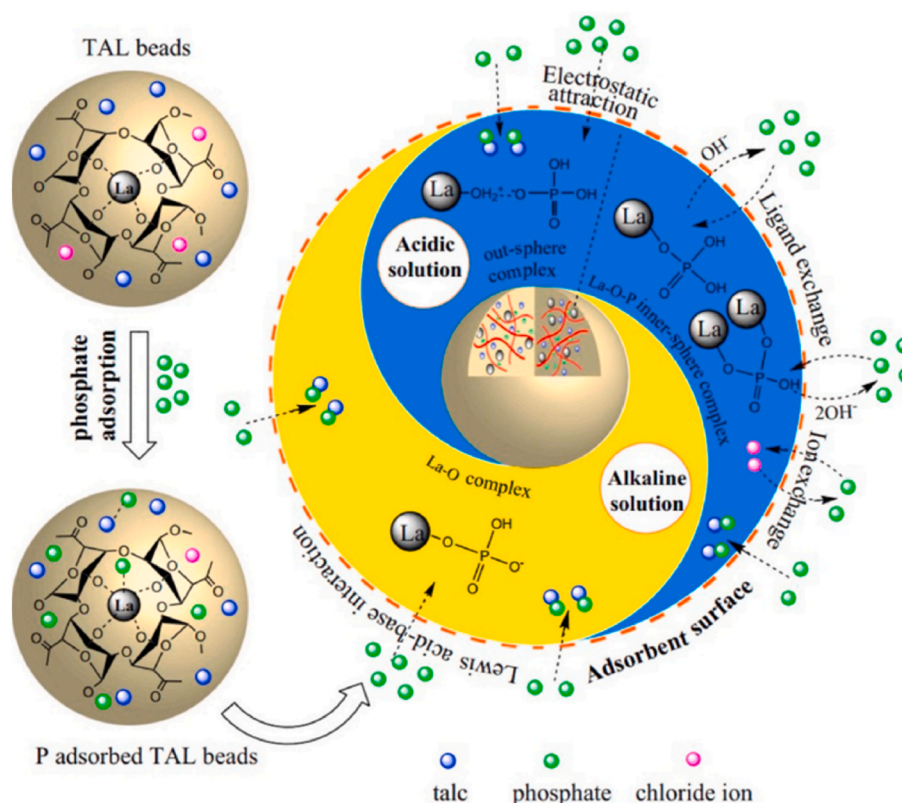


Fig. 9. Illustration of potential mechanisms for phosphate removal employing TAL. Reprinted (reproduced) with permission from (Wang et al., 2020), Elsevier.

properties make it valuable in paper, food, textiles, pharmaceuticals, cosmetics, and construction. Cellulose, a popular wastewater treatment adsorbent, is abundant, cheap, biodegradable, and non-toxic. Its hydroxyl groups interact with pollutants through ion exchange, adsorption, and chelation (Fakhri et al., 2023). Customizable for improved adsorption and selectivity, cellulose-mediated adsorbents effectively remove heavy metals, dyes, oil, organic solvents, pesticides, and pharmaceutical residues from wastewater (Xu et al., 2023). Cellulose exhibits exceptional heavy metal ion adsorption capability. Introduced a novel water purification method using grafted cellulose fiber named DMAE-PS (zwitterionic *N*-(3-sulfonato-1-propyl)-*N*, *N*-dimethylammonium) to remove Thallium (Tl) (Yang et al., 2023). DMAE-PS exhibited exceptional  $Tl^{+1}$  adsorption capacity (274.7 mg/g), outperforming other biomass-based materials. Notably, DMAE-PS also displayed high selectivity for  $Tl^{+1}$  over  $Zn^{2+}$ ,  $Cr^{3+}$ ,  $Mn^{2+}$ ,  $Cu^{2+}$ , and  $Cd^{2+}$ . The strong  $Tl^{+1}$  adsorption was attributed to a stable  $-SO_3^- \cdot Tl^{+1}$  interaction, which remained unaffected by fluctuations in pH (Fang et al., 2023).

Dyes are complex organic pollutants, that create global environmental challenges. Commonly used in industries like paper, printing, textiles, and cosmetics, they add color to products. Due to their widespread usage, dyes often end up in the environment through industrial wastewater, which is a big concern because large amounts of toxic dyes are discharged yearly (Khatooni et al., 2023). However, there is hope for tackling this problem, as PSA-mediated bio-adsorbents such as cellulose and its derivatives display promise in eliminating dyes from wastewater. For instance, utilized cellulose nanocrystals for this aim. (Soleimani et al., 2023) indicated that Cellulose nanocrystals were employed to create calcium alginate hydrogels, resulting in a powerful eco-friendly adsorbent for MB dye removal (max capacity: 676.7 mg/g) from wastewater. Based on the surface analysis, it was revealed that a rough and porous hydrogel prior to MB adsorption was formed. Moreover, MB adsorption led to hydrogel porosity loss, suggesting pore filling or diffusion as potential mechanisms.

On the other hand, (Khatooni et al., 2023) used a sodium

carboxymethyl cellulose-g-poly(acrylamide-co-methacrylic acid)/-Cloisite 30B nanocomposite hydrogel to remove MB from wastewater. The study indicated that the adsorbent had a surface area of  $149.61 \text{ m}^2 \text{ g}^{-1}$  and a max adsorption capacity of  $77.51 \text{ mg g}^{-1}$ . Ideal conditions for MB adsorption: pH 8, temperature  $45^\circ \text{C}$ , contact time 30 min, adsorbent dose  $0.8 \text{ g.L}^{-1}$ . Negative Gibbs free energy and enthalpy indicated spontaneous adsorption, boosted by high temperatures. The effectiveness of the nanocomposite hydrogel in eliminating Methylene Blue from wastewater relies on various factors, including the hydrogel's composition, dye concentration, contact duration, pH levels, and temperature (Hassanzadeh-Afruzi et al., 2022a; Mehdizadeh et al., 2022). Before large-scale implementation, comprehensive studies are needed to evaluate efficiency, cost-effectiveness, and environmental impacts.

Pharmaceutical industries have transformed healthcare, improved disease treatment and increasing global life expectancy. However, this progress presents challenges, like the release of high-concentration antibiotic waste into the environment (Mosavi et al., 2023). This wastewater from pharmaceutical plants and healthcare facilities can cause environmental problems: i) promoting antibiotic-resistant bacteria in the environment, making infections more difficult to treat with regular drugs (Gamoń et al., 2022); ii) harming beneficial microorganisms in soils and water bodies, disrupting ecosystems' balance (Rana et al., 2017); iii) posing health risks through exposure to antibiotic-contaminated water sources or agricultural products (Bielen et al., 2017); iv) prolonging the presence of antibiotics in the environment, exacerbating the aforementioned issues (Ding and He, 2010).

Traditional wastewater treatment processes may not be effective for removal of these complex and diverse compounds, leading to their presence in wastewater. Therefore, utilizing PSA-mediated bio-adsorbents for the adsorption of pharmaceutical pollutants in wastewater can be a major step towards mitigating the environmental impact of pharmaceutical industries and safeguarding water quality. By reducing the presence of these pollutants in aquatic systems, we can better protect ecosystems and human health. Recently, a highly effective eco-friendly

adsorbent was created to eliminate Ciprofloxacin and Levofloxacin antibiotics from wastewater. This polymer/clay nanocomposite hydrogel bead, made from carboxymethyl cellulose, acrylamide, and Fe clay via ionotropic gelation, achieved impressive removal rates of >92% for Ciprofloxacin and 93% for Levofloxacin under optimal conditions (Gopal et al., 2022). Therefore, integrating cellulose into biological nutrient removal systems would mitigate harmful concentrations of heavy metals, dyes, oil, and organic solvents that can harm microbial diversity and hinder biological nutrient treatment processes. It can be concluded that PSA-mediated bio-adsorbents play a significant role in wastewater treatment, removing nutrients, heavy metals, dyes, and pharmaceutical residues effectively. They also support microorganisms in biological nutrient removal systems. These bio-adsorbents offer eco-friendly solutions for wastewater treatment and hold promise for the future. Ongoing research aims to optimize their application in larger-scale systems, considering factors such as PSA type, nanomaterial, wastewater composition, and treatment process while assessing their environmental impacts. Achieving sustainable and efficient wastewater treatment practices relies on the effective use of these bio-adsorbents.

## 6. Concluding remarks

Although PSA has been widely used in fabrication of water purification membranes, a wider window of creativity has become opened by modification of PSA biopolymer. In recent decades, PSA-based nanocomposites have attracted the attention of researchers in both academic and industrial circles. Due to a host of favorable properties, which make them suitable choices for treatment/separation processes, they are poised to become sought-after biomaterials in a circular economy of the future.

The niche focus of this review was on the fabrication of membranes from PSA polymers reinforced with nanoparticles for deployment in wastewater treatment. Based on their provenance and microstructural properties, PSA-based nanocomposites emerge as decent candidates for use as adsorbents, photocatalysts, and membranes. Typically, process selection and technology choices for the removal of contaminants from wastewater depends on the initial capital investments, operation and maintenance expenses, the initial pollutant concentrations in the wastewater, safety concerns related to environment, pH level of wastewater, and more practically and economically depends on the required degree (efficiency) of removal. PSA-based adsorbents benefit biological systems by boosting microbial activity. Cellulose-mediated ones effectively remove pollutants (heavy metals, dyes, oil, solvents, pesticides, pharmaceutical residues, etc.) from wastewater. PSA nanocomposites aid wastewater treatment plants. The photocatalytic strategy is a unique method because it does not require any chemical supplements, resulting in reduced amount of precipitates that should be handled. PSA membranes reinforced with nanoparticles are another class of PSA nanocomposites used in water treatment.

## 7. Future perspectives and outlook

The future of water/wastewater treatment strategies based on PSA-based nanocomposites appears highly promising, offering a range of innovative and effective solutions to address the global challenge of water pollution. As research and development continue to progress, several key areas are growing to shape the outlook of PSA-based nanocomposites for efficient wastewater treatment:

1. The field of nanotechnology is continuously evolving, and further breakthroughs in material synthesis and functionalization will lead to the development of more efficient and versatile PSA-based nanocomposites. These advancements may enhance adsorption, photocatalysis, and membrane-based methods, enabling removal of a broader range of contaminants.
2. The successful implementation of PSA-based nanocomposites in real-world industrial settings, particularly in pharmaceutical and chemical industries, will become a priority in the near future. As the concern over antibiotic waste and other harmful chemical discharge grows, these industries will seek reliable and cost-effective solutions for sustainable wastewater treatment plants.
3. PSA-based nanocomposites have the potential to enhance biological processes in wastewater treatment plants. By effectively removing heavy metals and other pollutants that inhibit microbial activity, these nanocomposites can boost the overall efficiency of biological treatment methods, leading to cleaner water and reduced environmental impacts.
4. Researchers may investigate, evaluate and quantify the suitability of various PSA for wastewater treatment, identifying new and potentially more effective materials for nanocomposite fabrication. This could lead to the discovery of novel properties and functionalities, expanding the range of applications for PSA-based nanocomposites.
5. As the utilization of PSA-based nanocomposites increases, it becomes more and more essential to conduct comprehensive environmental impact assessments. Understanding the potential risks associated with the use and disposal of these nanocomposites would be crucial in ensuring their safe and sustainable deployment in wastewater treatment.
6. Transitioning from laboratory-scale experiments to large-scale applications will be a critical step in realizing the full potential of PSA-based nanocomposites for wastewater treatment. Overcoming challenges related to production, cost-effectiveness, and integration into existing infrastructure will be necessary to facilitate widespread adoption.
7. As PSA-based nanocomposites become more prevalent in the wastewater treatment industry, regulatory frameworks will likely be established to govern their use and ensure compliance with environmental standards. Clear guidelines and standards will aid in fostering responsible deployment and managing potential risks.

In the light of above, the future of PSA-based nanocomposites for water/wastewater treatment looks promising, with the potential to revolutionize the industry. Ongoing research and responsible implementation could help mitigate water pollution, safeguard public health, and preserve ecosystems globally (Table S5 in the SI). However, these all depend on management and budgeting, which are sometimes part of financial or political constraints on development. Moreover, diversity of PSA sources obtained from extraction of plants, vegetables and bio-wastes and biomass should be taken into account in classification of PSA for target application. Accordingly, bio-based nano-scale reinforcements to be added to PSA should be green, compatible with PSA, and economically justifiable. Therefore, practicality of proposals and lab-scale investigations should seriously be taken into consideration.

## Author contribution

Hussein E. Al-Hazmi: Writing- Review & Editing the first draft; Review & Editing the final version; Justyna Łuczak: Writing- Review & Editing the first draft; Investigation; Sajjad Habibzadeh: Review & Editing the final version; Supervision; Mohamed S. Hasanin: Writing- Review & Editing the first draft; Ali Mohammadi: Writing- Review & Editing the first draft; Review & Editing the final version; Amin Esmaeili: Writing- Review & Editing the first draft; Formal Analysis; Seok-Jhin Kim: Review & Editing the final version; Mohsen Khodadadi Yazdi: Writing- Review & Editing the first draft; Navid Rabiee: Review & Editing the final version, Supervision; Michael Badawi: Review & Editing the final version; Mohammad Reza Saeb: Review & Editing the final version, Supervision, Conceptualization.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

No data was used for the research described in the article.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2023.140578>.

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