Journal of Composites and Compounds 5 (2023) 159-178

Available online at www.jourcc.com

Journal homepage: www.JOURCC.com

Journal of Composites and Compounds

Advancements in nanoparticle-supported Laccase immobilization: Offering promising solutions for water treatment

Sogand Bahadori [a]*, *Maryam Azimpour* [b]

[a] *Department of Chemistry, Kerman Branch, Islamic Azad University, Kerman, Iran*

[b] *Hospital of Shahid Ayatollah Dastgheib, Shiraz University of Medical Science, Shiraz, Iran*

A B S T R A C T

Water purification has become a crucial matter in various societies. Increasing demand for water treatment is a constant challenge in the field of water treatment. Nowadays enzyme immobilization plays a crucial role in the realm of water cleansing. Also, laccase is the most common enzyme used in this context. Laccase is an enzyme that has gained popularity in various water treatment applications due to its ability to degrade organic pollutants and contaminants. It can be used in combination with different materials, including nanoparticles, to enhance its performance in water purification processes. Laccase is known for its effectiveness in breaking down a wide range of organic compounds, making it a valuable tool in the field of water refinement and environmental remediation. This study aims to delve into current knowledge about the role of nanoparticles supported in laccase immobilization for water purification. Our exploration method describes the use of metal-based, magnetic, carbon-based, and other nanoparticle supports along with novel methods including cross-linking, covalent binding, encapsulation, adsorption, and layer-by-layer assembly used in laccase immobilization. Results reveal the effectiveness of diverse nanoparticles in enhancing laccase stability and activity. In conclusion underscores the potential of nanoparticle-supported laccase immobilization as a sustainable solution for water treatment, offering improved enzyme performance and reusability.

A R T I C L E I N F O R M A T I O N

Article history:

Received 16 March 2023

Received in revised form 28 June 2023

Accepted 11 July 2023

Keywords:

Laccase immobilization

Nanoparticle-supported materials

Metal-based materials

Magnetic nanomaterials

Carbon-based nanomaterials

Immobilization methods

Water treatment

Table of contents

* Corresponding author: Sogand Bahadori; E-mail: Sogolbahadori@gmail.com

https://doi.org/10.61186/jcc.5.3.2

1. Introduction

Laccases, also known as benzenediol: oxygen oxidoreductase (EC 1.10.3.), represent a type of extracellular multicopper enzyme within the blue copper oxidases category. This group of enzymes is notable for their unique ability to facilitate the one-electron oxidation of phenolic compounds, coupled with the simultaneous reduction of molecular oxygen to form water. Remarkably, laccases achieve this oxidation process without relying on hydrogen peroxide. Their substrate specificity is generally modest, permitting them to perform oxidation on various substances, such as o- and p-diphenyls and phenols containing methoxy substitutions [1].

Laccase immobilization has significant promise as a prospective 'eco-friendly' biocatalyst for environmental uses, such as remediating contaminated soil and treating wastewater [2]. Laccase immobilization technology holds immense importance for environmental sustainability, while research into its application in water purification brims with energy. First and foremost, this synthesis outlines the attributes of immobilization methods and carriers, elucidating their impact on laccases. Furthermore, it unveils the mechanisms behind pollutant removal facilitated by immobilized laccases, shedding light on the intricate interactions among pollutants, carriers, and laccases subsequently, The utilization of immobilized laccase to enhance the process of water purification [3]. Considering that the stability of immobilized enzymes hinges on their interaction with the carrier, utilizing nanoparticles as carriers emerges as a viable choice due to their substantial surface area-to-volume ratio [4]. The process of laccase immobilization brings forth numerous enhancements, such as safeguarding its activity and elevating its stability beyond that of free laccase. Furthermore, the capacity to reuse immobilized laccase stands as a notable advantage with potential applications in the future [5].

There have many studies on these nanoparticles, some studies present carbon-based nanoparticle antimicrobial activity, and demonstrate the significance of carbon nanoparticle size in deactivating microorganisms. Numerous investigations have been conducted within the realm of nanoparticles. Certain studies have unveiled the antimicrobial properties exhibited by carbon-based nanoparticles, emphasizing the significance of carbon nanoparticle size in deactivating microorganisms [6]. Furthermore, other scholarly examinations have delved into various categories of CNMs (carbon nanomaterials) like carbon nanotubes [7], reduced graphene oxide(rGO), graphene oxide(GO), graphene, carbon black, graphite, and fullerene [8].

Characteristics, manufacturing methods, benefits, hurdles, and forthcoming pathways for magnetic nanoparticles [9]. All in all, applications, properties, demerits, merits, and other aspects of these Nanoparticles were the ultimate goal of past research. The techniques employed to affix the enzyme onto the carriers assume a pivotal role in determining the performance of the immobilized enzyme. Diverse immobilization methods encompass Layer-by-Layer Assembly, Cross-Linking, Encapsulation, Covalent binding, Covalent binding, Entrapment and physical Adsorption. Several methods were scrutinized, including Efficiency, Application, biocatalyst properties, and Stabilization. The aim was to lay the groundwork for selecting suitable support for laccase immobilization in future studies [10]. Other research delved into laccase immobilization within polymers through various techniques: Entrapment, encapsulation, covalent binding, cross-linking, and adsorption. These methods were analyzed for their impact on pollutant removal performance, along with an examination of their pros and cons [11]. Adsorption relies on hydrophobic interactions, hydrogen bonds, and Van der Waals forces for immobilization, offering benefits such as simplicity, enzymatic activity retention, and reversibility. Encapsulation utilizes polymeric micelles with bio-polymeric supports, achieving high immobilization yields and stability [12]. Nano-based technologies show great promise in optimizing water treatment processes, enhancing efficiency, and reducing costs [13]. Nanoparticles find application in the elimination of contaminants, wastewater treatment, and aiding Nanofiltration, offering solutions to combat the issues presented by waterborne pollutants and microorganisms [14]. The objective of this review study is the current state of knowledge regarding methods for laccase immobilization using nanoparticles to explore their applications in water treatment. This explores the latest advancements in laccase immobilization facilitated by nanoparticles, emphasizing their transformative role and potential applications exclusively in water treatment.

2. Nanoparticle-Supported Laccase Immobilization

Laccase (EC 1.10.3), a common multi-copper oxidase derived from higher plants, insects, and microorganisms, can catalyze the oxidation of diverse organic compounds by utilizing molecular oxygen as the electron receptor, resulting in the production

of water as a by-product [15]. However, the practical industrial applications of free laccase are inhibited because of their low stability and non-reusability as well as high production cost [15]. However, the poor reusability and stability of free laccase have limited its large-scale applications [16]. Nevertheless, laccase would be denatured in extreme pH, temperature metal ions conditions, and organic solvents. A plausible solution is to immobilize enzymes on specific materials and surfaces [17]. Enzyme immobilization is a proven method for reusing efficient catalysts, product separation, and recycling [18]. Immobilization of laccase into polymers, [19] metal-organic frameworks (MOFs)[20], and various nanoparticles were seen in different research. Recently, nanoparticles have been widely reported in the enzyme immobilization field, owing to their unique properties such as surface-to-volume ratios, [17] readily available on a commercial scale, chemically modified to create appropriate functional groups on the surface for enzyme attachment, adjustable to preferred sizes with large surface areas, altered for improved biocompatibility, possessing significant rigidity to maintain stability during immobilization, and exhibiting magnetic properties for convenient separation from the reaction mixture using a magnet compared to non-magnetic supports [21]. Physical adsorption, covalent binding, encapsulation, and cross-linking are different strategies employed to immobilize enzymes [22]. Different types of nanoparticle-supported to immobilize enzymes are shown in Figure 1.

3. Type of nanoparticles used in laccase immobilization

3.1. metal-based nanoparticles

Over recent years, there has been a growing interest in using metal-organic frameworks (MOFFs) for enzyme immobilization. These MOFs are quite intriguing because they are a unique type of porous material made by linking metal-based nodes and organic ligands. What makes them special is their precisely arranged crystal, roomy pores, and large surface area [23]. Metal-organic frameworks (MOFs) consist of metal-organic and node ligands linked through coordination bonds. The arrangement and characteristics of MOFs can be modified based on their various metal nodes and ligands, allowing for the management of the interplay between enzymes and MOFs. In contrast to conventional enzyme carriers immobilized through uncontrolled pore sizes, MOFs offer advantages including lower production expenses, reduced enzyme leaching, and weak product stability [23]. The metal-organic framework not only secures the enzyme in place, safeguarding its sturdy structure,

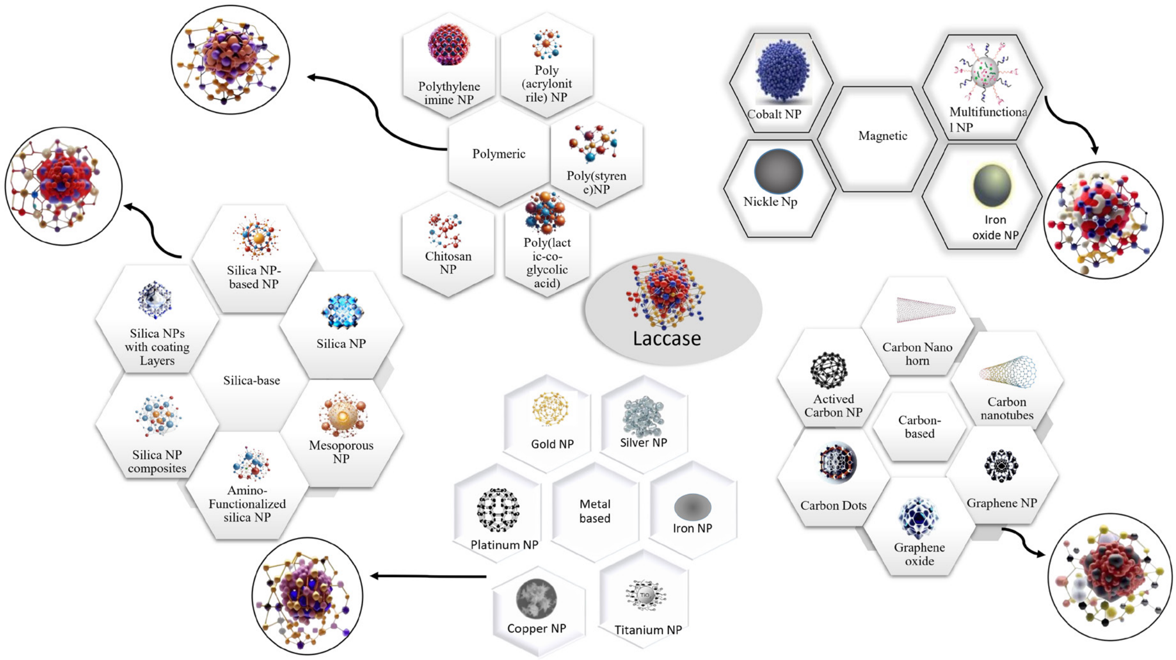

Fig. 1. Immobilized laccase with diverse types of Nanoparticle-supported.

but it also facilitates full contact between the enzyme and the substrate [24]. Various types of metal-based nanoparticles exhibit biocidal properties, including titanium dioxide (TiO), Copper-based, zinc oxide (ZnO), silver (Ag) and other lesser-known metals such as platinum (Pt), nickel (Ni), silicon dioxide (SiO), gold (Au), cerium oxide (CeO) and iron-based The observed behaviors of metal nanoparticles related to their antibacterial effects and potential mechanisms of action against bacteria have been emphasized [25]. Metallic nanoparticles, along with polymeric materials containing distinct functional groups [7], find application in the alteration of these nanoparticles to enhance their stability. The introduction of metal nanoparticles elevates the conductivity of nanocarbons, ultimately enhancing the efficiency that allows them to combat bacteria that have already become resistant. Furthermore, they engage with a multitude of biomolecules, which disrupts the formation of antibiotic-resistant strains of Direct Electron Transfer (DET) [26]. Predominantly inorganic nanoparticles based on metal materials, stand out as highly favored particles within the realm of inorganics. They offer a hopeful response to the challenge of growing antibiotic resistance. These nanoparticles don't simply rely on mechanisms of action akin to traditional antibiotics; instead, they employ entirely distinct approaches. This unique mode of action allows them to combat bacteria that have already become resistant. Furthermore, they engage with a multitude of biomolecules, which disrupts the formation of antibiotic-resistant strains [27].

In soil, heavy metal ions have the potential to assail the active center of FL (free laccase), leading to a decline in laccase stability. Immobilization, in contrast, bestows laccase with improved resistance to harsh conditions and thermal stability when compared to its free form. This enhancement enables laccase to be reused, delivering numerous benefits for environmental applications [5, 28]. Enzyme synthesis involves implementing metal-based nanostructures on a large scale. The magnetic nature of metallic nanoparticles, which allows for high recyclability, positions them exceptionally among nanomaterials used for enzyme immobilization. Various functional groups contribute to enhancing biocatalytic immobilization. Metallic nanocarriers, prized for their exceptional properties and stability, have emerged as promising options for immobilizing enzyme additives in immobilizing phytase. The enzymatic activity witnessed a remarkable 5% increase upon the immobilization of probiotic cells. This enhancement resulted from the heightened cationic density of chitosan due to the chelation process, leading to increased teichoic acid and zinc absorption onto B coagulants. Furthermore, the chitosan-ZnO immobilized cells exhibited improved thermostability characteristics and PH [29, 30].

Metal-organic frameworks (MOFs) constitute a category of porous crystalline materials, comprised of metal-based nodes and organic ligands featuring various functional groups such as carboxylates, amines, nitrates, phosphates, and sulfonates. These materials amalgamate the advantageous attributes of both inorganic and organic substances. Using a range of post-synthetic modification techniques, MOFs' chemical properties can be meticulously customized to suit particular applications through a thoughtful approach to designing their nodes [29, 31].

Metal-based nano adsorbents offer a cost-effective solution for efficiently eliminating heavy metal pollutants from water. Nanoparticles like ferric oxide, titanium oxide, manganese oxide, and magnetic materials have undergone extensive research and proven their potential for treating effluents [32]. Their large surface-to-volume ratio makes them more effective than traditional sorbents. Furthermore, the pH of the nanoparticle solution significantly influences their interaction with heavy metals, enhancing the advantages associated with metal-based nano adsorbents. For instance, they are less toxic, possess substantial surface areas for interactions, and exhibit chemical distinctiveness, rendering these metal oxide nano adsorbents even more appealing and unique [23, 33].

Laccase, an enzyme used for oxidizing organic compounds, faces stability issues in specific environments. Improving its stability and reusability is crucial for the industry. Immobilizing laccase on metal-organic frameworks (MOFs), and porous materials enhances its utility. MOFs offer an ideal platform due to synergistic interactions with laccase's metal ions. Challenges include MOF size constraints, requiring a deeper understanding of laccase changes. To maximize MOFs for laccase, further research, and innovative approaches are needed, addressing size issues, understanding structural changes, and optimizing MOF use [34, 35].

Copper-based nanomaterials, exemplified by copper-nucleotide/DNA coordination compounds, have undergone development to replicate laccase activity. Other nanomaterials with metallic foundations, such as platinum nanoparticles and manganese oxides, have also exhibited laccase-like characteristics. Regarding laccase immobilization, it stands as a pivotal endeavor for enhancing stability and reusability. Nonetheless, the quest for an economical carrier that preserves enzyme functionality remains a persistent challenge. Endeavors have been initiated to fashion enzyme mimics, commonly referred to as nanoenzymes, to surmount the limitations associated with native laccase and immobilized variants. Nanozymes bear several advantages, including cost-effectiveness, scalability in production, extended shelf-life, robust stability, and adjustable reactivity [36, 37].

Qingqing Wang et al. [38] in their article mention that metal-based supports have been receiving attention for laccase immobilization. It states that metal-chelated enzyme adsorption is based on interactions between metal ions on the support and certain groups in the enzyme. Various metal-based supports such as Cu-, Ca-, Al-, and Zr-based supports have been studied for laccase immobilization, and they have shown improved laccase activity and stability.

About advantages of metal-based supports in the article highlights that metal-based supports offer advantages for laccase immobilization. It mentions that metal-chelated enzyme adsorption is relatively simple and less expensive compared to other methods. Metal-based supports, such as electrospun nanofibers, are recognized as excellent supports for enzyme immobilization due to their large surface area, high recovery, and reusability of the enzymes. Polyacrylonitrile (PAN) nanofibers for laccase immobilization The article specifically mentions that PAN nanofibers, after surface modification, have been studied and proven to be an ideal support for laccase immobilization. It states that amidoxime polyacrylonitrile (AOPAN), prepared by the chemical modification of the nitrile group in PAN is known as a good adsorbent for metal ions. The AOPAN nanofibrous membranes chelated with different metal ions, including Fe3+, Cu+, Ni+, and Cd+, were successfully used as carriers for laccase immobilization.

Dawei Li et al. [39] showed zeolitic imidazolate framework-90 (ZIF-90) was utilized to encapsulate laccase (LAC), resulting in the formation of ZIF-90/LAC biocomposites. These biocomposites, in combination with bacterial cellulose (BC) and carboxylated multi-walled carbon nanotubes (c-MWCNTs), led to the development of a novel cellulose membrane with remarkable biocatalytic

properties. Acting as a biosensor electrode, the membrane exhibited a linear response to catechol within the range of 20 to 400 μM, showcasing a low detection limit of 1.86 μM (S/N = 3), alongside exceptional selectivity, reproducibility, and stability. Furthermore, in water treatment applications, the membrane surpassed pure LAC in catechol degradation efficiency, maintaining a degradation range of 93.4% to 82.1% over five cycles. This innovative membrane presents significant promise for monitoring and effectively treating phenolic wastewater, highlighting its versatile potential in environmental remediation.

Metal-based nanoparticles, such as gold, silver, and copper, exhibited varying surface areas and biocompatibility profiles. Gold nanoparticles demonstrated a higher surface area and moderate biocompatibility, while silver nanoparticles displayed a comparatively lower surface area but better biocompatibility. Copper nanoparticles showed moderate surface area and biocompatibility, providing insights into their potential for enzyme binding in laccase immobilization [27, 40].

The diverse properties observed among metal-based nanoparticles underscore their suitability for enzyme immobilization. Gold nanoparticles, with their higher surface area, may offer more binding sites for enzyme attachment, while silver nanoparticles, despite the lower surface area, present better biocompatibility. Copper nanoparticles, exhibiting moderate properties, could offer a balanced compromise for laccase immobilization strategies. The choice of metal-based nanoparticles can be tailored based on specific requirements, balancing surface area, biocompatibility, and binding efficiency [41, 42].

3.2. Magnetic nanoparticles

Magnetic nanoparticles (MNPs) have emerged as a versatile tool in various fields, offering unique properties that make them ideal candidates for enzyme immobilization and other bioactive applications. Magnetic nanoparticles (MNPs), particularly those derived from magnetite Fe_3O_4, have garnered significant interest due to their compatibility, minimal toxicity, and robust magnetic characteristics, with biological substances [43].

Immobilizing Enzymes Using MNPs is the wide array of applications for laccase and the benefits of Fe_3O_4 as a magnetic material for enzyme immobilization have sparked the exploration of fresh approaches to immobilize laccase, expanding its potential in various industrial sectors [35].

MNs made of Fe_3O_4 have been considered as favorable substrates due to their precisely defined surface characteristics, reduced mass transfer impediments and expansive surface areas. Enzymes anchored onto Fe_3O_4 magnetic nanoparticles can not only uphold their distinctive functionality but also facilitate effortless separation and recyclability [44].

The use of magnetic nanoparticles as a support for enzyme immobilization has opened up new possibilities for employing solid biocatalysts. This approach allows for the retrieval of enzymes, extends their applicability in continuous processes, and provides safeguarding against thermal and chemical alterations during storage or manufacturing [45].

The advantages of Magnetic bio-separation technology represent a promising approach for enzyme immobilization support systems. It enables swift separation and straightforward retrieval in an external magnetic field, all while reducing both initial investment and operational expenses [46].

Diverse applications of MNPs offer a wide array of uses, extending beyond just enzyme immobilization. They possess advantages such as a substantial surface area, robust mechanical properties, ease of chemical modification, and distinctive magnetic characteristics. These properties make them valuable in applications like biological imaging, and immunosensing detection. Magnetically targeted drug delivery, and more [47].

About MNs for the Elimination of Heavy Metals is said magnetic nano adsorbents made of Fe_3O_4 exhibit magnetic characteristics that simplify separation, even from viscous solutions. They are employed in the extraction of metallic elements such as Pb+ and Cr_6+ [48].

Efficient Removal of dyes using magnetic nanoparticles offers highly effective capabilities for the elimination of dyes from water sources [49].

Magnetic thermal and optical properties of NPs rely on diverse elements, hence it is essential to thoroughly assess their physicochemical characteristics to improve their effective utilization [45].

The magnetic property enables effortless separation from the reaction mixture using a magnet, a capability absent in non-magnetic supports. Numerous valuable enzymes, such as cellulose, lipase, laccase, and dehydrogenase, find extensive application in both industrial and environmental sectors [21].

Matrices of magnetic nanoparticles are highly efficient and versatile tools for sample preparation, offering: Efficient Extraction, Quick Separation, Customizability, Selectivity, Reusability, and Reusability. In short, magnetic nanoparticle matrices are fast, efficient, customizable, and eco-friendly for various sample preparation needs [50]. Porous nanostructures offer superior surfaces for enzyme entrapment or covalent binding, but the key challenge lies in designing and synthesizing structures with ideal surface properties and biocompatibility.

Magnetic nanocomposites are created through sol-gel techniques, chemical precipitation methods, or the NanogenTM microwave plasma method, combining magnetic nanoparticles and biopolymers [51]. Figure 2 shows different morphologies of magnetic nanocomposite materials

Efforts have focused on developing carrier-bound immobilized enzymes for use in continuous processes, especially considering cost constraints. Magnetic nanoparticles (MNPs) have emerged as promising candidates due to their non-toxic, biocompatible, and magnetic properties. In recent years, there has been a growing interest in MNPs and their applications, including drug delivery, hyperthermia treatment, cell separation, biosensors, enzymatic assays, biocatalysis, and environmental remediation. MNPs with various surface modifications are used for immobilizing enzymes such as yeast alcohol dehydrogenase and lipase. For experiments, glucose oxidase (Gox), a flavoprotein with dual active sites, catalyzes the oxidation of β-D-glucose to gluconic acid while reducing oxygen to hydrogen peroxide. Gox serves as a model enzyme for evaluating different immobilization techniques and has significant potential in biosensors and biofuel cells [52, 53].

Cristiano C.S. Fortes et al. [43] focused on the optimization of laccase immobilization on magnetic nanoparticles (MNPs) for biocatalytic reactions. MNPs are considered suitable supports for enzyme immobilization due to their ability to be easily separated from reaction media using an external magnetic field. The immobilization process was optimized using a box-Benhken experimental design, resulting in the successful binding of laccase to functionalized MNPs. The immobilized laccase showed improved thermal stability and retained above 75% of its activity after 6 consecutive cycles of reaction. The study highlights the potential of using MNPs for laccase immobilization, offering benefits such as reusability and cost reduction.

Sanjay K. S. Patel et al. [54] presented the covalent binding of RvLac onto magnetic nanoparticles FeO_3 and Fe_3O_4, which were modified with APTES and subsequently treated with glutaraldehyde. The immobilization of RvLac was more effective on FO_3 nanoparticles due to their smaller size and larger surface area in comparison to Fe_3O_4 nanoparticles. Following immobilization, the enzyme displayed enhanced activity at elevated pH levels and temperatures, along with notably improved stability compared to its free form. The RvLac immobilized on FeO_3 particles exhibited sustained high reusability and demonstrated increased degradation of bisphenol A. Prior studies have focused on RvLac immobilization using non-magnetic supports. This newly developed biocatalyst based on magnetic nanoparticles exhibits promising potential for various biotechnological applications beyond bisphenol A degradation.

Somayeh Mojtabavi et al. [55] laccase immobilized on hercynite ($FeAl_2O_4$) magnetic nanoparticles (MNPs) demonstrated remarkable efficacy in removing ciprofloxacin from hospital wastewater (HWW) with enhanced stability and efficiency. The hercynite MNPs, synthesized through co-precipitation, were surface-modified with copper (II) ions to facilitate coupling with enzyme functional groups. The laccase@ Cu^2 + @hercynite•MNPs maintained 50% of its initial catalytic activity after 13 cycles of reuse and exhibited significantly improved stability compared to free laccase during storage at 4 °C and 25 °C. The immobilized laccase effectively removed 85% of ciprofloxacin from HWW within 3 hours at 40 °C, employing p-coumaric acid. The removal mechanism involved defluorination, hydroxylation, and piperazine ring cleavage, reducing antibiotic toxicity against both Gram-positive (G+) and Gram-negative (G–) bacteria. Consequently, the study suggests that laccase@Cu^2 + @hercynite•MNPs hold promise for micro-pollutant bioremediation in hospital effluents, Emphasizing their potential as an efficient tool for addressing antibiotic residue concerns in wastewater treatment.

Diana C Sotelo et al. [56] Examined five magnetic biofilters integrating magnetic nanoparticles (142 nm) and laccase immobilized on nanoparticles (190 nm), alongside permanent magnetic elements like neodymium magnets and metallic meshes. Assessing Congo Red dye decolorization, filter longevity, and losses of magnetic nanoparticles and enzymes revealed that filters with laccase-immobilized magnetite, permanent magnets, and metallic meshes achieved the highest Congo Red decolorization (27%) and a seven-cycle lifespan. Dye decolorization varied from 5% to 27% across different filter types. Despite greater magnetite losses in magnet-containing filters (57 mg), the use of permanent magnetic elements notably tripled the filter's lifespan compared to those lacking enzymatic properties and doubled the lifespan compared to laccase-magnetite filters, showcasing a promising advancement in wastewater treatment.

Magnetic nanoparticles showcased unique characteristics conducive to laccase immobilization. Surface modifications improved their enzyme binding efficiency. Their magnetic nature allowed easy separation from reaction mixtures using a magnet, enabling efficient enzyme reuse [57].

The distinct magnetic properties of these nanoparticles offer significant advantages in enzyme immobilization processes. Enhanced enzyme binding efficiency and the ability to be easily retrieved post-reaction, ensuring repeated utilization, make magnetic nanoparticles a promising choice for laccase immobilization. Their magnetic nature facilitates their recovery, making them highly attractive for scalable industrial applications [58, 59].

3.3. Carbon-based nanoparticles

Carbon-based nanomaterials, including carbon nanotubes (CNTs), graphene, and others, have garnered substantial attention across various scientific disciplines owing to their extraordinary properties. Exploration of the diverse characteristics and applications of these substances [60].

In recent years, nanocarbons, a category of carbon-based nanomaterials, have emerged as groundbreaking materials with distinct attributes. These nanomaterials boast exceptional properties that find applications across a wide array of domains, solidifying their status as indispensable elements in contemporary technology and science [61].

Carbon nanoparticles, renowned for their versatility, have been employed not only in immobilizing enzymes but also in various applications. These applications encompass even coatings, energy storage, solar cells, and microelectronics. This stems from their remarkable thermal and electrical conductivity, as well as their impressive tensile strength. The adaptability of carbon nanoparticles in various industries is further highlighted by their wide range of uses [62].

The Examination and Classification of Carbon-Based Nanomaterials encompass thorough investigations into a variety of carbon-based nanomaterials, involving nano horns nanodiamonds, graphene, fullerenes, carbon nanotubes, carbon nanocones/disks, carbon nanofibers, and fullerenes. These nanomaterials have been methodically categorized into four distinct groups, guided by their inherent properties and emerging patterns: carbon nano horns, carbon nanocones/disks, Nano diamonds, fullerenes, carbon nanofibers, carbon nanotubes a graphene [63].

The captivating field of research that involves manipulating molecules at the nanoscale draws from a wide array of components. Materials like carbon, boron, TiS, MoS, NbS, WS, chrysotile (asbestos), kaolinite, and other source materials are used to craft nanostructures, including fullerenes and nanotubes. When compared to their bulk counterparts these nanostructures exhibit exceptional electronic physical and mechanical properties [64].

The properties of carbonaceous nanomaterials encompass their electronic, chemical, and physical. Aspects, intricately depend on carbon's structural arrangement and hybridization. It becomes crucial to comprehend the foundational orbital setup of carbon, characterized by six electrons distributed across 1s, s, and p orbitals. A critical factor lies in the slim energy separation between the s and p electron shells, enabling the elevation of one suborbital electron to the vacant higher-energy p orbital, a phenomenon absent in the ground state [64, 65].

Carbon-based nanoparticles have garnered widespread usage within environmental systems, particularly in the realm of analytical appli-

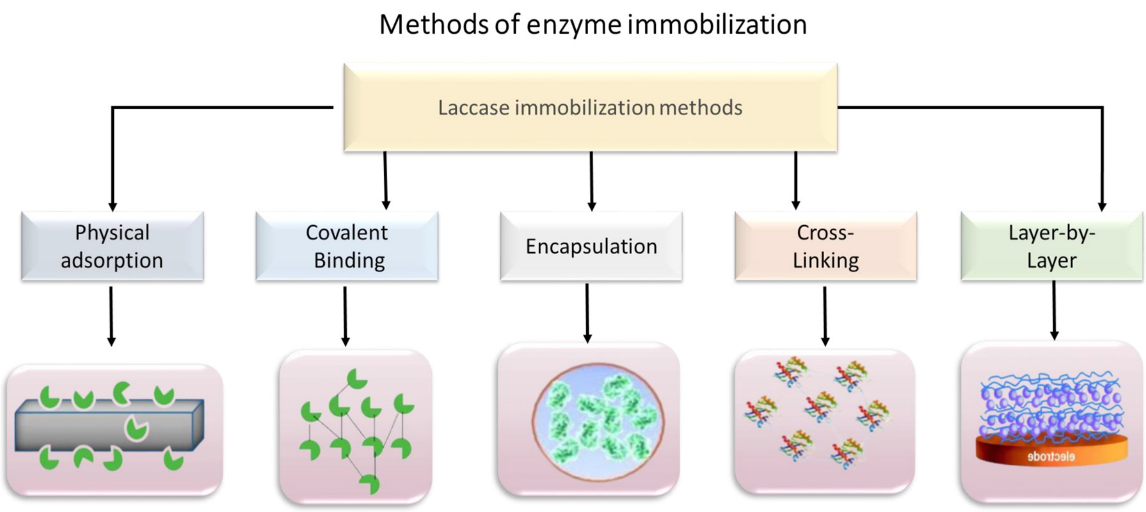

Fig. 2. Various matrix types for magnetic NP.

cations. The selection process between various carbon allotropes can sometimes lack precision, often hinging on past material availability and experiences. Nevertheless, an extensive spectrum of carbon-based substances has been implemented in the field of analytical procedures [66].

Addressing the Issues and Improving the Dispersion and Solubility of Carbon-Based Nanoparticles, which encompass a range of materials like nanotubules, nanorings, nanofibers, peapods, nano onions, and nanodiamonds, can at times face challenges due to the presence of strong van der Waals' forces. Various pre-treatment methods have been suggested by researchers to mitigate these hindrances. For instance, incorporating polar groups like oxygen, hydroxyl, polyvinylpyrrolidone, and phenyl has been proposed. Nonetheless, it's worth noting that these approaches can also influence stability and other aspects, including electrical, optical, mechanical, and magnetic properties [67, 68]

Furthermore, the atom plane reactivity of carbon-based nanomaterials is different from those situated at the periphery. Explorations have been conducted regarding the electrochemical characteristics of atoms found at the edge-plane locations of CNTs. these investigations have revealed similarities to the behavior observed in various planes of graphite. It's also noteworthy that the presence of non-metallic and metallic impurities within CNTs have demonstrated an impact on electrochemical activity [69].

Regarding the adsorption capacities and selectivity of Carbon Nanotubes (CNTs), they pique substantial interest because of their distinct cylindrical form, often with dimensions below 1 nm. CNTs may take the form of solitary or multi-layered structures, each endowed with individual attributes. The notable surface area, substantial porosity, and hollow structure of CNTs, inclusive of external grooves, and selective properties, collectively bestow upon them remarkable adsorption capacities, interstitial gaps, and inner locales [70, 71]

Functionalization through various treatments, such as acid introduction and the incorporation of functional metals or groups, further amplifies their adsorption capabilities. CNTs have exhibited outstanding performance in the capture of pollutants such as mycotoxins, heavy metals, and other impurities from aqueous solutions, rendering them invaluable in endeavors aimed at environmental remediation [72].

The growing significance of environmental reclamation can be attributed to the distinct characteristics and diverse range of carbon-based structures. Various scientific domains, consisting of engineering, physics, and chemistry. Have experienced a surge in ground-breaking advancements and opportunities, all propelled by the material's adaptability [73].

Carbon-based materials have demonstrated their effectiveness in eliminating a wide spectrum of contaminants from water sources. These pollutants encompass pesticides, noxious metal ions, metalloids, pharmaceuticals, and a variety of both inorganic and organic substances. Carbon-based adsorbents, including carbon nanotubes and graphene, have emerged as prevalent choices for water purification. The efficiency of a carbon-based adsorbent in the removal of chemical compounds relies on the specific adsorbate and the conditions of the solution [74, 75].

In the realm of enzyme immobilization using Carbon-Based Nanomaterials, one of the significant domains of application for nanomaterials derived from carbon lies in the process of enzyme immobilization. Materials such as (CNTs), Graphene, and their counterparts exhibit remarkable potential within this sphere. Their outstanding attributes, including excellent thermal conductivity, resistance to high temperatures, chemical inertness, and compatibility with biological systems, render those prime candidates for the immobilization of enzymes [76].

Nevertheless, it's essential to acknowledge that employing a single carbon-based nanomaterial for enzyme immobilization might come with certain limitations. These encompass suboptimal material resilience, challenges in achieving dispersion, and, at times, reduced effectiveness. Researchers are actively tackling these issues to further augment their applicability [57, 77]

The extensive usage of carbon nanomaterials extends to laccase immobilization, showcasing their versatility in this application. The success of laccase immobilization hinges on several factors, consisting of the chemical surface area, the presence of functional groups on these materials' surfaces, and chemical composition. Carbon-based substances like activated biochar, graphene, and carbons have appeared as top contenders for the immobilization of laccase [78].

Their intricate presence, high surface area, and pore structures of various functional groups render them highly desirable options for this purpose. Researchers are actively exploring the potential of these materials for efficiently immobilizing laccase enzymes, and the results obtained so far are promising [79].

Avinash A. Kadam et al. [80] discuss the use of carbon-based materials like carbon black and carbon nanotubes to immobilize laccase, enhancing its biosensing capabilities. Various immobilization techniques are explored, including adsorption, entrapment, cross-linking, and covalent bonding. These techniques involve interactions like hydrophobic and hydrogen bonding for laccase attachment. Covalent immobilization, while affecting laccase structure, enhances its specificity for phenolic compounds.

Nano-immobilization supports are deemed crucial for laccase-based biosensors. The article highlights modifying nano-supports like silica nanoparticles and multi-walled carbon nanotubes with functional groups. It also suggests using nanostructures that mimic laccase catalysis (nanozymes) for future biosensor development.

In summary, the article underscores the importance of carbon-based materials, diverse immobilization techniques, and nano-supports in enhancing laccase-based biosensors' performance.

Luffa sponge-based magnetic carbon nanocarriers: The document introduces luffa sponge as a renewable biomass resource with a special porous structure and high carbon purity. It suggests that luffa sponge can be used as a raw material for the preparation of magnetic carbon nanocarriers (MLCs) through a one-step carbonization-magnetization process. These MLCs can be used for laccase immobilization and have shown good magnetic properties and a strong load capacity for laccase. Application in bisphenol A removal: The document mentions that the immobilized laccase on luffa sponge-based magnetic carbon nanocarriers (Laccase@MLC-1) showed superior catalytic performance compared to free laccase in the degradation of bisphenol A (BPA). Laccase@MLC-1 exhibited stronger thermal stability, better acid tolerance, and higher BPA degradation efficiency. It was able to completely remove 100 mg/L of BPA in 4 hours, while free laccase was only removed [81].

Michaela Patila, et al. [82] discussed the utilization of carbon-based nanomaterials for laccase immobilization, with a focus on their effectiveness for environmental and industrial applications. They synthesized hybrid nanomaterials by combining smectite nanoclays with carbon-based materials like GO, carbon nanotubes, and adamantylamine [83]. This approach successfully immobilized laccase from Trametes versicolor (TvL) with high yields, and the immobilized TvL retained its activity even after prolonged exposure to high temperatures, unlike the free enzyme which became inactive. Furthermore, the immobilized TvL on carbon-based nanomaterials exhibited exceptional decolorization capabilities, making it suitable for efficiently degrading synthetic dyes. Importantly, this immobilized enzyme demonstrated remarkable reusability, performing well for up to 11 successive catalytic cycles. The article primarily focuses on the immobilization of TvL to decolorize phenolic dyes using these innovative hybrid nanomaterials. This study provides valuable insights into the application of laccase immobilization on carbon-based nanomaterials as an eco-friendly and effective approach for dye degradation processes.

Roopkumar Sangubotla, et al. [84] in their article discusses the use of carbon dots (CDs) for the immobilization of laccase enzyme. The CDs were synthesized using curcumin and dimethylformamide and then functionalized with a silicon precursor called 3-(aminopropyl)-triethoxysilane (APTES). The resulting APT-CDs were used as a platform for laccase immobilization. The laccase enzyme was covalently immobilized onto the APT-CDs to create a novel bioprobe. The immobilization process involved the addition of glutaraldehyde (GA) to the APT-CDs, followed by rinsing with phosphate-buffered saline (PBS) and laccase solution. The resulting bioprobe showed a colorless powder appearance and was refrigerated for further experiments. Overall, the article highlights the use of carbon-based materials, specifically CDs, for the immobilization of laccase enzymes. The immobilized laccase bioprobe showed fluorescence properties and was successfully immobilized on a tapered optical fiber for the detection of dopamine.

Mahsa Masjoudi, [85] laccase immobilization onto a polyvinylidene fluoride (PVDF) membrane modified with multi-walled carbon nanotubes (MWCNTs) was investigated for the elimination of pharmaceutical pollutants, carbamazepine, and diclofenac. The covalently immobilized laccase from Trametes Hirsuta showcased remarkable activity (4.47 U/cm2) and a noteworthy activity recovery of 38.31%. Notably, the immobilized laccase demonstrated improved operational and thermal stability compared to its free form. Utilizing immobilized laccase in a mini-membrane reactor led to substantial removal efficiencies, achieving 27% removal within 48 hours for carbamazepine and an impressive 95% removal in just 4 hours for diclofenac. The findings underscore that laccase immobilized on PVDF/MWCNT membranes serves as a promising catalyst for large-scale water and wastewater treatment, offering compatibility with existing treatment facilities while effectively addressing pharmaceutical contaminant removal.

Wenxiang Zhang et al. [86] Illustrated laccases were directly immobilized onto carbon nanotubes (CNTs) to enhance their adsorption capabilities for removing recalcitrant micro-pollutants in wastewater. Comprehensive investigations compared adsorption performance between carbon nanotubes and laccase-carbon nanotubes under various operational conditions. Results showed laccase-carbon nanotubes achieved a higher dye removal rate (96% within 3 hours) and better stability than carbon nanotubes (84% within 3 hours) across diverse parameters, such as carbon nanotube concentration (0.02–0.08 g/L), laccase ratio (0.25–1.25), dye concentration (10–60 mg/L), temperature (15–35 °C), and rotating speed (0–250 rpm). Analyses indicated laccase-carbon nanotubes possessed greater adsorption capacity and faster diffusion rates for dye removal, potentially due to timely dye elimination by laccases, allowing regeneration of adsorption capacity. The study presents a promising biomimetic nanocomposite for improving wastewater treatment methods targeting stubborn micro-pollutants.

Various carbon-based nanoparticles, including graphene and carbon nanotubes, exhibited distinctive characteristics relevant to laccase immobilization. Graphene showcased high surface area and carbon nanotubes displayed exceptional structural properties, both contributing to enhanced enzyme binding capabilities. However, challenges related to dispersibility and aggregation were observed, impacting their practical use [57, 87].

Carbon-based nanoparticles, especially graphene and carbon nanotubes, demonstrate promising attributes for laccase immobilization. Their high surface areas offer abundant binding sites, potentially improving enzyme binding efficiency. Despite their advantageous properties, issues regarding dispersibility and aggregation pose challenges in practical applications, necessitating further research to address these limitations for scalable utilization [35, 88]. Some immobilized laccase with carbon-based NP supported is shown in Figure 3.

3.4. Silica and other nanoparticles

There are extensive diverse of nanoparticles for Laccase Immobilization, various nanoparticle categories encompass Composite hydrogels, self-assembled 3D, and Graphene/polymer. Polymers exhibit a wide range of mechanical characteristics, and hydrophilic, chemical, and structural attributes, making them a versatile tool for the immobilization of laccase [89].

Regarding materials selection for enzyme immobilization researchers have undertaken extensive efforts to choose various substances as supporting materials, ranging from traditional options like alginate, natural polymers, microporous resin, and glass, diatomite, to a variety of nanomaterials (materials with nanostructures) [90].

Related to Silica-Based Nanomaterials relevance it can be mentioned that Silica-based nanomaterials e.g., MCM-$_{41}$, MCM-$_{48}$, SBA-$_{15}$ exemplified by silica SiO find extensive application in the immobilization of enzymes due to their attributes, including a high specific surface area, structured pores, controllable pore size, and morphology, excellent stability, non-toxicity, and a surface amenable to functionalization. Among these materials, mesoporous silica (SiO) composed of an inorganic silicon framework (Si–O–Si), stands out as the most thoroughly explored and mature material to date [91].

Diverse Support Materials for Enzyme Immobilization turn back to Silica and its derivatives stand out as the most frequently utilized

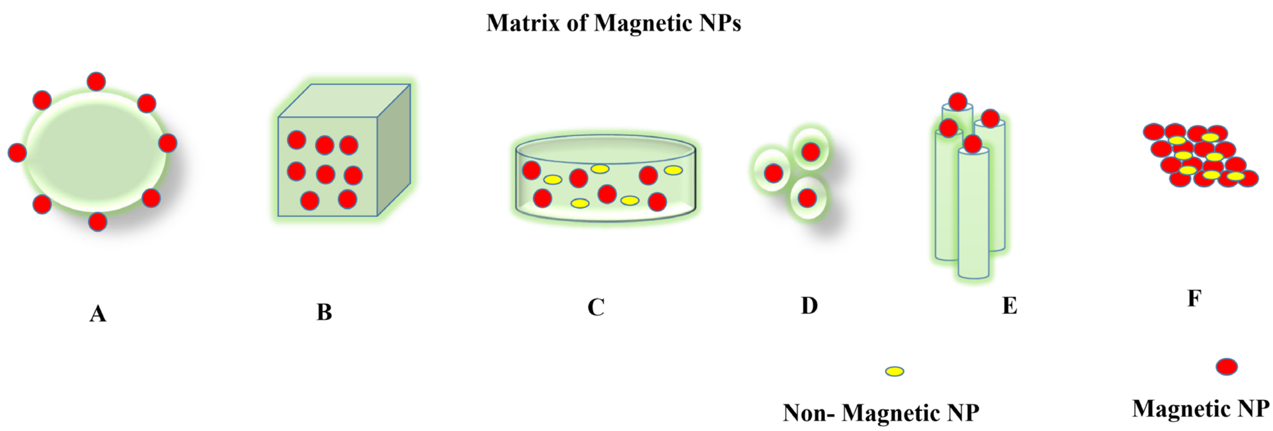

Fig. 3. Immobilized laccase supported by Carbon-based NPs.

substances for supporting enzyme immobilization. The hydrophilic nature of the silica surface, along with the abundance of hydroxyl groups, enables the immobilization of biomolecules through adsorption, covalent bonds, and even encapsulation[92]. Various materials of organic origin have also been employed as supports for laccase immobilization, including both natural and synthetic polymers, utilizing a range of immobilization techniques. Organic materials can also be engineered with controlled porosity. Additionally, magnetic iron oxides II and III inorganic metal oxides like titania, alumina, and magnetic iron oxides have found widespread use in laccase immobilization. These materials offer a substantial surface area, allowing for increased enzymatic loading and reduced mass transfer resistance to substrates [10].

For improved Removal and Stability of Pollutants, the application of laccase onto fumed silica nanoparticles was found to enhance the enzyme's long-term stability, enabling the effective removal of bisphenol-A and sodium diclofenac from wastewater [93].

Silica-based solid support is highly effective for immobilizing biomolecules like enzymes, proteins, and DNA across diverse applications, including biosensors and interfacial studies. The modification of silica surfaces has provided valuable insights. Combining nanoparticle advantages with silica's surface modification potential holds promise for nanoscale biosensors. However, there's limited exploration of nanoparticle-based biosensors. Polymeric nanoparticles in biochemistry face challenges like self-aggregation in biological settings and wide size variations, hindering their effectiveness in precise analytical measurements [94].

Immobilized laccase exhibits elevated stability concerning temperature, pH, storage, and reusability, For instance, some studies noted broader pH and temperature ranges for immobilized laccase, coupled with significantly improved 2,4,6-trichlorophenol (TCP) removal (95.4%) compared to free laccase. Other studies demonstrated the efficiency of immobilized laccase: Chitosan-clay composite magnetic microspheres achieved 80% bisphenol A (BPA) removal in 4 hours, while the removal efficiency of 2,4-dichlorophenol (DCP) reached 87.6% compared to 82.7% for free laccase under optimal conditions. These findings highlight the substantially improved efficiency of immobilized laccase compared to its free form in water treatment applications [3].

Polymeric nanoparticles, often abbreviated as NPs, encompass particles sized between 1 to 1000 nanometers. They possess the capacity to contain active substances either trapped within or adhered to the surface of the polymeric core. The designation "nanoparticle" encompasses both nanocapsules and nanospheres, distinguished by their morphological structure [95].

Modified Polymeric membranes to enhance enzyme reusability, and ultrafiltration membranes made of polymers or polysulfone serve as commonly adopted carriers for enzyme immobilization through physical adsorption. This approach notably enhances the ability to reuse enzymes when removing micropollutants. It's essential to emphasize that the percentage of materials (whether nanomaterials or enzymes), aspect ratio and the size of the carrier are pivotal factors within the realm of membrane technology that requires meticulous consideration [96].

Recent investigations in the field of biocatalytic membrane technology have focused on the study of various nanoparticles. These nanoparticles include graphene oxide nanosheets, titanium dioxide, nickel–zinc magnetic nanoparticles, silica-based and Fe_3O_4 chitosan nanoparticles, and materials utilized for enzyme immobilization techniques [97].

To achieve enzyme immobilization, mesoporous silica-based nanosupports rely on physical absorption techniques, including electrostatic hydrophilic and hydrophobic linkages. The regulation of enzyme immobilization varies depending on the specific enzymes, with mesoporous silica nanoparticles playing a crucial role. They do so by modifying surface characteristics, such as the presence of distinct functional groups, surface charge density, and pore geometry [89].

In certain research discussions, it shelled explore the process of laccase immobilization. They have carried out experiments to secure laccase in place with the help of artificial polymers, there are in a wide array of sizes, ranging from the nanoscale to the macroscale. Cellulose and chitosan stand out as two frequently utilized polymers in this context, and their roles in eliminating pollutants have been prominently featured. This can be attributed to the fact that they possess functional groups capable of adsorption, thereby facilitating straightforward modifications. Multiple techniques, including cross-linking, encapsulation, and entrapment, have been employed for this specific purpose. Also, they delved into the impact of laccase immobilization on various operational parameters, including temperature [98].

Mesoporous silica-based nano-supports rely on physical absorption methods, including electrostatic, hydrophilic, and hydrophobic linkages, to carry out the immobilization of enzymes. The adjustment of surface properties such as pore geometry, surface charge density, and the presence of specific functional groups plays a crucial role in regulating the immobilization of diverse enzymes by mesoporous silica nanoparticles [99].

Yao Zhu et al. [100] developed a modified poly(vinylidene fluoride) membrane (PVDF) with high mechanical strength and chemical stability for laccase immobilization using covalent bonding. The synthesis involved constructing a hybrid bio-inorganic structure on a polydopamine (PDA)-coated PVDF surface by grafting 3-triethoxysilylpropylamine (APTES) modified Fe_2O_3@SiO_2 cubes (FS@cubes) through a solvothermal process. This process resulted in the formation of FS@cubes-PDA@PVDF membranes, onto which laccase was immobilized via glutaraldehyde (GA) crosslinking (Lac-FS@cubes-PDA@PVDF). The Lac-FS@cubes-PDA@PVDF exhibited a remarkable 97.1% removal efficiency of Congo Red under optimal reaction conditions (pH 7.0 and temperature 35℃), surpassing the efficiency of free laccase. Furthermore, the prepared Lac-FS@cubes-PDA@PVDF displayed excellent stability after low-temperature storage and remarkable reusability, offering a potential strategy for water pollutant removal and a convenient approach for large-scale enzyme-catalyzed applications.

Graphene-based nanomaterials (GNPs) exhibit an enhanced capability to capture a wide spectrum of chemicals. However, they also come with a notable drawback: the challenging separation of adsorbed substances from the original solution. To address this issue, nanomaterials like magnetic nanoparticles (MNPs) are integrated into reduced (rGO) or (GO) sheets. MNPs simplify the process of separating the adsorbent from the initial solution. Fe3O4 serves as an exemplary MNP, possessing a substantial surface area, high reactivity, minuscule particles, and non-toxic properties. These characteristics contribute to their benefits and elevate their desirability for such applications [101]. Composites and nanohybrids based on graphene exhibit outstanding chemical, mechanical, electrical, and thermal characteristics. Consequently, they are considered promising options for the applications and immobilization of enzymes in the field of nanomedicine. Various methods for enzyme immobilization have been proposed and explored in recent years. However, the most suitable approach depends significantly on factors such as the specific enzyme being used and its commercial and practical viability [102]. Graphene is vital in the realm of enzyme biofuel cells (EBFCs) because of its sustainability. The presence of both SP and SP3 orbitals within graphene is what makes it possible for direct electron transfer (DET), a critical factor for improving EBFC performance. The incorporation of (rGO) and its derivatives into EBFC construction stands as a strategic maneuver, enabling the seamless facilitation of DET between the enzyme laccase and the electrode. This strategic synergy ultimately culminates in the attainment of maximum power output [103].

Feng Wang, et al. [104] in their article discuss the immobilization of laccase, an enzyme, on mesoporous silica nanoparticles. The nanoparticles have a large-pore wormhole framework structure, which provides highly accessible reaction sites for enzymatic catalysis. The immobilization of laccase on these nanoparticles was achieved through metal affinity adsorption using a chelating agent. The immobilized laccase showed higher adsorption capacity and activity recovery compared to physical adsorption. The immobilized laccase also exhibited improved storage stability and temperature endurance. The method used in the paper involved synthesizing large-pore magnetic mesoporous silica nanoparticles using tetraethyl orthosilicate as the silica source and amine-terminated. Jeffamine surfactants as templates. The nanoparticles were functionalized with iminodiacerate through a silane-coupling agent and chelated with Cu+. This functionalized nanoparticle, called MMSNPs-CPTS-IDA-Cu+, was used for the immobilization of laccase through metal affinity adsorption. The immobilized laccase showed high adsorption capacity, activity recovery, and improved stability and endurance.

In other research Jelena Bebić et al. [105] focused on the immobilization of laccase from Myceliophthora thermophile on amino-modified fumed nano-silica (AFNS) nanoparticles, with a focus on optimizing the process and its potential application in lindane degradation. The study investigates various factors affecting immobilization, finding that the highest specific activity is achieved at an optimum pH of 5.0. The methodology involves using AFNS nanoparticles and enzyme solutions in a buffer, varying enzyme concentrations, pH, and immobilization time.

The results indicate that laccase immobilization occurs through adsorption as a monolayer enzyme binding, and the highest specific activity and immobilization yield are achieved at pH 5.0 and with an offered protein concentration of 160 mg per g of AFNS. The immobilization process occurs rapidly in 40 minutes, following pseudo-first-order kinetics. This study provides valuable insights into the immobilization of laccase on AFNS and its potential application in bioremediation, particularly for lindane degradation.

Saeed Kashefi, [106] (GO) was synthesized using a modified Hummer's method and utilized as an optimal support for enzyme immobi-

Table 1.
Summary of nanoparticle-supported laccase immobilization for water treatment

Type of NP	Characteristics	Properties	Advantages	Disadvantages	Application in water treatment	Ref.
Metal-based NP	Gold, silver, or platinum. Exhibit high catalytic activity	Small size, high surface area, excellent electron transfer.	Excellent efficiency, and enhanced stability.	Limited reusability, and cost.	Remove organic pollutants, phenolic compounds, and dyes.	[124] [125] [126]
Magnetic NP	Iron oxide core, magnetic. Easily manipulated with magnets.	Magnetism, superparamagnetic, high surface area	Easy separation, potential agglomeration.	Remove heavy metals and organics in magnetic processes.	Effective removal of heavy metal ions like lead and arsenic from contamination.	[127] [128] [129]
Carbon-based NP	Carbon nanotubes and graphene. Large surface area and high conductivity.	Greater enzyme loading, and good electron transfer.	Effective for pesticides, pharmaceuticals, may need functionalization.	Degrade pesticides, pharmaceuticals, and emerging contaminants.	Effective degradation of organic pollutants, such as pesticides and pharmaceuticals	[130] [131] [131]
Silica NP	Porous, Biocompatible, Stable. High surface area.	Stable microenvironment, controlled release.	Leaching risk, biosensing capabilities.	Biosensing of pollutants, removal of organics.	biosensing applications, removal of organic contaminants from water	[132] [133] [134]
Polymeric NP	Biodegradable polymers, tailored. Tunable properties	Biodegradability, tunable, Protection.	Limited stability under extremes.	Targeted removal of contaminants like oil, and microorganisms.	targeted removal of contaminants and water remediation processes	[116] [135]

lization due to its unique chemical and structural properties. Laccase from genetically modified Aspergillus was covalently immobilized onto GO, forming a nanobiocatalyst. Enzymatic characterization revealed significant parameters: laccase loading reached 156.5 mg g−1 with an immobilization yield of 64.6% at a laccase concentration of 0.9 mg/ mL. Employing various structural characterization techniques confirmed the morphological properties of the nanomaterials, including FTIR, XRD, SEM, TGA, and TEM. Investigating the bioconversion of anionic dyes (Direct Red 23 and Acid Blue 92) using the nanobiocatalyst demonstrated over 75% average decolorization effectiveness for both dyes over six cycles, highlighting its exceptional operational stability and efficient reusability in water treatment applications.

Silica nanoparticles exhibited significant surface area and demonstrated high stability in laccase immobilization. Their porous nature facilitated efficient enzyme binding, offering promising stability and enhanced reusability. However, considerations regarding potential cytotoxicity and limited functionalization for enzyme binding were noted [107, 108].

Silica nanoparticles present a favorable option for laccase immobilization due to their high stability and porous structure, facilitating efficient enzyme binding [109].

While their stability and potential for enzyme reuse are advantageous, concerns regarding cytotoxicity and limited functionalization for optimal enzyme binding need careful consideration for broader industrial applications [110].

Further advancements in surface modification may mitigate these concerns, making silica nanoparticles more versatile for enzyme immobilization [111].

In the field of water treatment, selecting the most suitable nanoparticles depends on the specific requirements and the type of pollutants present [112, 113]. However, carbon-based nanoparticles, such as carbon nanotubes and graphene, tend to be highly favored due to their extensive usage and remarkable attributes [114].

These carbon-based nanoparticles boast several beneficial traits for water treatment: Ample Surface Area: They possess a substantial surface area, enabling effective adsorption of contaminants [115, 116].

Increased Loading Capacity: Their unique structure allows for a higher loading capacity of enzymes or other treatment agents [117, 118].

Versatility: They are versatile and capable of effectively treating various pollutants like pesticides, pharmaceuticals, and emerging contaminants [119].

Although other nanoparticle types, such as metal-based (e.g., gold, silver, platinum), magnetic (e.g., iron oxide), silica, and polymeric nanoparticles, offer their advantages, carbon-based nanoparticles stand out due to their exceptional adsorption abilities, extensive surface area, and versatility in handling a broad spectrum of water pollutants [120, 121].

As a result, there is no definitive answer to which nanoparticle is the best for laccase immobilization in the water treatment field. Different nanoparticles have different advantages and disadvantages depending on the type of pollutants, the immobilization method, and the operating conditions. However, some studies have suggested that carbon-based nanoparticles, such as graphene oxide, carbon nanotubes, and activated carbon, are promising candidates due to their high surface area, func-

Table 2.
Summary of efficiency and stability nanoparticle-supported laccase immobilization.

Category	Used Nanostructure	Efficiency	Stability	Ref.
Metal-based NP	platinum nanoparticles (Pt) loaded within the crystalline metal-organic framework (MOF) ZIF-67	94.5%	Composite Material Reusability Suggests Stability	[136]
	Platinum nanoparticles (Bio-Pt) and palladium nanoparticles (Bio-Pd) synthesized with Desulfovibrio vulgaris	94%,85% and 70%	-	[137]
	Zerovalent copper nanoparticles (Cu np)	63% and 68%	Characterization techniques support stable integration.	[138]
Carbon-based NP	Laccase-loaded Pomegranate Peel-derived activated carbon(LMPPs0	69.8%	-	[139]
Magnetic NP	Iron Magnetic Nanoparticles (iMNP)," and "Composite Magnetic Nanoparticles(cMNP).	90%	-	[140]
	superparamagnetic Fe_3O_4 nanoparticles	95%	95%	[141]
	(MNPs) stabilized with poly-allylamine-hydrochloride (PAAH)	99.48%	Safe biocompatible drinking water.	[142]
Silica-based	silica-based ceramic hollow fiber	99%	CHFM/CRHA stable up to 1400°C.	[143]
	porous silica-based nano-adsorbents	99.5% and 99.4%	was evident in their ability to maintain high biodegradation efficiencies even when the temperature was increased to 37°C and when the samples were shaken at 150 rpm	[144]
Polymeric NP	Fe_3O_4 @SiO_2@PNP-SMIP	99%	-	[145]
	polymeric ultrafiltration membrane with embedded nano-silver particles	96.1% and 87.3%	95.0%	[146]

tional groups, and aromatic structure [145]. Carbon-based nanoparticles can also enhance laccase production by acting as supports, nutrients, and inducers in the culture media [78]. However silica-based NPs have shown good efficiency in the field of water treatment in comparison with carbon-based NPs, carbon-based NPs have some advantages and disadvantages for laccase immobilization that are some of the main points based on Zhang et al. [145] and Saptashwa Datta et al. [146] Silica-based nanoparticles are widely used as supports for laccase immobilization due to their high surface area, chemical stability, biocompatibility, and easy functionalization.

However, they also have some drawbacks, such as low mechanical strength, high leaching rate, and low enzyme loading capacity [147]. On one hand, some studies have reported that carbon-based nanoparticles have higher laccase Immobilization efficiency and catalytic activity than silica-based nanoparticles for the degradation of certain pollutants, such as dyes and phenols. This may be due to the higher affinity and interaction between laccase and carbon-based nanoparticles as well as the synergistic effect of redox Mediators.

One possible reason for the preference of carbon-based nanoparticles over other nanoparticles for laccase immobilization in water treatment is that carbon-based nanoparticles can act as redox mediators, which can enhance catalytic degradation of pollutants by laccase Redox mediators are small molecules that can transfer electrons between laccase and the substrate, thus expanding the substrate range and increasing the reaction rate[3]. Carbon-based nanoparticles such as GO and carbon nanotubes,

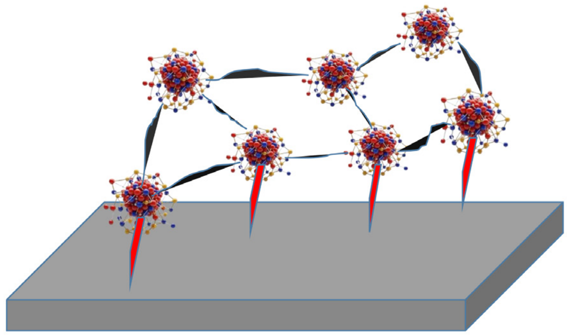

Fig. 4. Schematic picture of immobilization methods examined in this part.

Table 3.
Summary of the various nanoparticle-supported laccase immobilization methods and their applications.

Type of Nanoparticles	Immobilization Methods	Application	improved properties	Ref.
Carbon-Based NP	physical Adsorption, covalent bonding	industrial processes, Repeat reaction.	increases the pH, operating temperature, Operational stability.	[172]
	physical Adsorption	water pollutant treatment, dye decolonization, PHA removal.	good separation capacity, novel functional features, and good Reusability, high loading capacity.	[147]
	physical Adsorption	micro-pollutant removal, diffusion resistance, saturation reduction	exhibit synergy effects promoted equilibrium	[152]
Magnetic NP	Encapsulation Crosslinking Adsorption	Industries, textile, pharmaceutical, food processing, removal of pollutants, synthesis of fine chemicals, enzymatic transformations	Enhanced Stability Reusability Improved Catalytic Efficiency Environmental Friendliness	[173]
	cross-linking	Increases activity, Reused multiple times, reducing costs.	enhances activity exhibits stability good reusability	[174]
	Covalent Bonding cross-linking	enhance enzyme efficiency and stability	enhanced activity, stability, substrate degradation, storage stability, magnetic responsiveness, and tolerance to alkaline pH and high temperature	[175]
Metal-Based NP	Entrapment, adsorption, cross-linking, and covalent bonding.	Enhances concentration, and stabilization of enzymes, leading to improved biosensor performance.	Development of aerometric biosensors, optimizing the construction.	[176]
	Physical Adsorption	biodegradation, biosensing, and organic synthesis,	exhibit excellent catalytic, ideal supports, repeatability	[20]
	Physical Adsorption	co-immobilization remarkable biosensing and biocatalysts	higher optimal temperature, sustainable activity	[177]
Silica-Based NP	cross-linking	efficient catalytic activity, water purification potential	cost-effective, Enhances their stability and facilitates, overcoming limitations.	[178]
	Adsorption	Support for the enzyme, good biocompatibility, ease of separation, and ease of reusability.	increased enzyme activity, Enhanced Substrate Binding	[179]
Polymeric NP	Layer-by-layer assembly	controllable enzyme layers, convenient and efficient	Increased power density, enhances electron transfer, controllable immobilization, and improved stability.	[180]
	Layer-by-layer assembly	Creating novel matrix, cellulose support	Enhancing toughness, and conductivity, suitable for biosensors and biofuel cells.	[181]

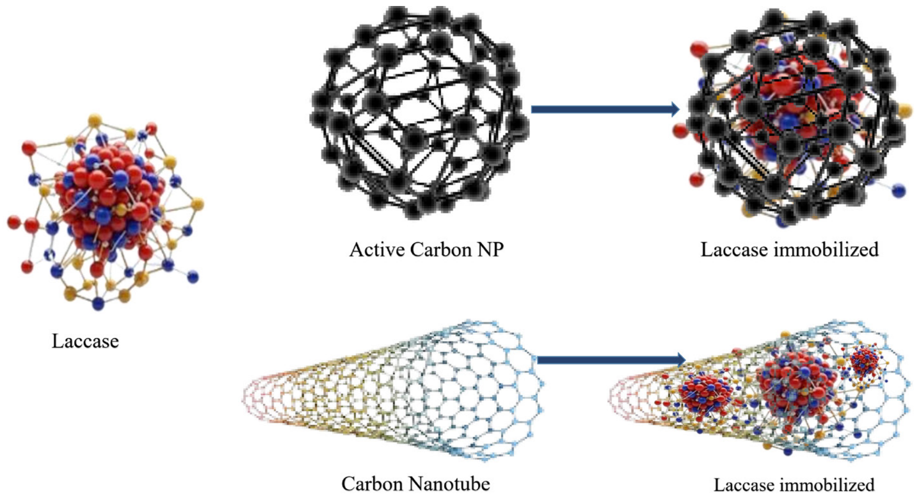

Fig. 5. Covalent binding method for laccase immobilization.

have been shown to exhibit redox mediator-like behavior, which can improve the efficiency of laccase immobilization and its application in water treatment [78]. Therefore, carbon-based nanoparticles can offer a dual function of supporting laccase and mediating electron transfer, which makes them more favorable than other nanoparticles that lack this property.

4. Methods for Nanoparticle-Supported Laccase Immobilization

Several methods have been developed for nanoparticle-supported laccase immobilization, including physical adsorption, covalent binding, and encapsulation. Physical adsorption involves the non-covalent binding of laccase to the surface of nanoparticles through electrostatic interactions, hydrogen bonding, or van der Waals forces. This method is simple and cost-effective but may result in low enzyme loading and poor stability. These methods enhance enzyme stability and activity, enabling their efficient utilization in various biotechnological applications, including wastewater treatment and biofuel production. Figure 4 shows several methods for nanoparticle-supported laccase immobilization.

4.1. Physical Adsorption

Carriers and laccase are bound with different binding forces making ion or physical adsorption. The adsorption method has some advantages such as repeatable use, convenient operation as well as low operation [3].

In physical adsorption, the preservation of enzyme conformation largely stems from the fact that adsorption primarily occurs through either van der Waals' forces or electrostatic interactions. However, due to the relatively weak nature of these bonds, enzyme detachment from the support material can occur during operation [148].

Fullerenes, nonporous carbon, carbon nanotubes, and graphene are carbon-carbon nanomaterials used as carriers in the physical adsorption method of enzyme immobilization. According to Skoronski et al. [149] and Zhang et al. [145] studies, such nanomaterials have much more enzyme loading capacity related to great mechanical strength, large surface area, and high electrical conductivity of Nano matters.

Zhou et al. reported a greater adsorption rate and higher adsorption capacity by using an enzyme for the removal of micro-pollutant adsorbed on (GO) surface. Degrading the pollutant by enzyme, releasing the preoccupied active points, and delaying the adsorption saturation level have been cited as the reasons behind the results [150].

4.2. Covalent Binding

Covalent immobilization methods are crucial in enhancing laccase stability by preventing enzyme leakage into the reaction mixture [151]. This aspect is particularly vital for industrial applications where enzyme activity needs to be maintained over extended periods [152]. The process of covalent bonding allows for precise functionalization of supporting materials, enabling tailored modifications to enhance the binding efficiency between the enzyme and the support matrix[153]. This customization can significantly influence the stability and activity retention of the immobilized enzyme [154].

The interaction between enzymes and their supporting materials categorizes immobilization methods into two primary groups: physical methods, which include entrapment, encapsulation, and adsorption, and chemical methods, like self-immobilization and covalent binding onto solid supports. Among these methods, covalent bonding stands out as particularly intriguing for industrial applications. Covalent immobilization not only enhances enzyme stability but also prevents the leakage of enzymes into the reaction mixture [106]. Figure 5 shows immobilized laccase with covalent bonding method.

Covalent bonding might introduce limitations in substrate transfer due to strong enzyme-support binding, potentially leading to alterations in enzyme conformation and an increase in Km values, affecting the enzyme's affinity towards its substrate [155].

For immobilization of laccase onto GO Nanosheets by covalent bonding, Skoronski et al. [149] first applied the nitration process of GO, then used sodium borohydride for oxidation and reduction reactions and finally added glutaraldehyde for crosslinking. Greater operational activity retention has been demonstrated by a covalent binding method in comparison with the adsorption process. However, both adsorption and covalent binding methods showed high stability of the enzyme in the rough conditions of temperature and pH.

The covalent immobilization of laccase on magnetic NPs resulted in a higher Km compared to free enzymes, primarily due to the strong binding of the enzyme to the support, causing limitations in substrate transfer or undesired conformational alterations. The immobilized Cu/FeO4-laccase, for bisphenol degradation, displayed higher pH (4.0) and temperature (45°C) optima compared to the free enzyme [21].

Mogharabi-Manzari et al. [119] used magnetic mesoporous silica spheres as enzyme-support matter and physical covalent binding as the immobilizing method. The findings showed high resistance against various pH, great stability in thermal changes, and activity retainment compared to free laccase.

4.3. Encapsulation

Among methods to immobilize enzymes, the application of the adsorption strategy is easy although its weak attachment caused enzyme leaching during operations. Covalent immobilization caused enzyme stability improvement, but deactivation of enzyme relatively occurs. Another technique such as enzyme incorporation in silica matrices has been shown to improve the enzyme efficiency as a catalyst. For instance, sol–gel is a matter to encapsulate the laccase used as a controlled release of bioactive compounds, an optional coating for electrochemical and optical biosensors, stationary phases for affinity chromatography, and immunosorbent and solid-phase extraction materials[156]. Laccase was encapsulated within a hydrogel created from a sol-gel mixture of TMOS and MTMS, following Veum et al.'s method for sol preparation. After this preparation, the mixture was cooled to 0°C, diluted with water, and used immediately for enzyme encapsulation. Laccase, initially dissolved in a 0.1 M phosphate buffer at pH 7, was then immobilized using different protein concentrations, leading to gelation within a few minutes. The resulting hydrogel was transformed into a powdered form, underwent washing, and was stored at 4°C. Additionally, aliquots of pH 7 phosphate buffer were introduced to protect against oxidation [157].

To encapsulate enzymes on different composites such as MOFs, paying attention to the molecular size of the enzyme is important. For example, enzymes with small molecular sizes like cutinase, micro peroxidase as well as cytochrome C (CytC) are suitable to encapsulate into MOFs composites compared to the laccase [13]. In other words, there are different structures of laccase caused by various molecular weights between 50 to 70 kDa. So, entering the laccase into most materials is difficult [20]. Laccase effectively immobilized within a hydrogel formed by a sol-gel matrix comprising TMOS and MTMS, exhibited exceptional reusability, retaining 59.8% activity even after ten cycles of use; MOFs were employed to enhance stability, albeit with the caveat that excessive encapsulation may impede enzyme activity in environmental exposure, while the enzyme's immobilization in Cu (PABA) was achieved using the coprecipitation method with the addition of methanol, thereby concluding the complex process [11].

The laccase encapsulation within the chitosan-nano biochar matrix represents an innovative approach for practical biocatalyst applications, where nano biochar serves as an effective pollutant adsorption support, facilitating prolonged contact time for degradation by the immobilized laccase; this study aimed to evaluate the performance of this immobilized biocatalyst system in terms of removal efficiency, enzyme stability, and recyclability, examining properties like stability under different pH and temperature conditions, potential enzyme reusability, antibacterial activity, leakage, and storage time, with the encapsulation method optimizing Laccase immobilization on Chitosan-Nano biochar [158] [159].

4.4. Cross-Linking

To overcome the free enzyme usage obstacles, the immobilization of enzymes by cross-linking enzyme aggregates (CLEAs) method is suggested. This method is a carrier-free strategy that improves enzyme productivity for the following reasons; carrier elimination, and multi-layer enzyme immobilization [13]. However, there are some restrictions in their utilization because of the dependence of a cross-linking procedure on Lys groups of enzymes. In other words, enzymes with a low number of Lye groups will produce weaker cross-linking attachment leading to detaching enzymes during reactions [160]. In the research conducted by Liu et al. [161] laccase was immobilized onto polyethyleneimine-magnetic Fe_3O_4 nanoparticles through Cu+ chelation. Activity retention and its loading were 91.65% and 5.19 mg/g, respectively. The stability of free laccase and immobilized laccase were compared. The results showed a significant improvement in the thermal and storage stability of immobilized laccase. Laccase activity maintained up to 51.45% under a storage temperature of 60 °C and time of 6 h and 81.13% under a storage temperature of 3 °C and time of I month.

A study showed laccase immobilized onto the surface-modified nanoparticles of Fe_3O_4 through the glutaraldehyde cross-linking method achieved 7% recovery in its activity and could maintain its initial activity after using in six consecutive cycles more than 30% [162].

4.5. Layer-by-Layer Assembly

Although covalent binding caused high enzyme stability because of enzyme-support immobilization, enzyme activity diminishes because of naturally changed enzyme structure. Therefore, the Layer-by-Layer (LBL) assembly method can be used to improve the mentioned defect and to maintain the enzyme activity as well as to increase enzyme stability. In addition, according to Sarma et al. research, reloading the enzyme on supported matters will be possible [163].

In the research carried out by Li et al. [164] the Acknowledged Layer-by-Layer (LBL) assembly method was applied to immobilization of laccase into the skin layer of the NF membrane to remove bisphenol.

The LBL assembly method is used to immobilize enzymes in enzymatic biofuel cells (EBFCs). By using nanomaterials, the power of the density of LBL assembly-based EBFCs heightens. In physical adsorption, as an immobilization method of enzyme, enzyme adsorbs onto porous materials. In this method enzyme's activity remains unchanged. On the other hand, a crosslinking method to immobilize enzymes produces a good stability attachment to cross-linkers, it requires complicated operations, though. Furthermore, electrochemical polymerization is another strategy used to immobilize enzymes on

the electrode. In this strategy, the reaction of polymerization is done by monomer molecules on the electrode under a specific external potential. Over the electrochemical polymerization process, enzymes acting as intermediates interact with the main chain of the polymer. Enzymes are attached to the electrode while molecules of the monomer are polymerized on the electrode surface. Because of directly capturing enzymes into a polymer such as sol–gel, mesoporous carbon, conductive polymer, etc. in the embedding method, the enzyme structure is kept unchanged and its redox reaction is maintained stable. LBL assembly is a kind of method to immobilize enzymes with a three-dimensional (3D) multilayer structure showing easier operability and better activity. By using the LBL assembly method, enzyme loading onto the surface of the electrode is increased [18]. Table 2 illustrates reviewing different methods for Laccase immobilization and their applications in recent research.

Efficient enzyme immobilization offers benefits like reusing enzymes, improving stability, easy reaction control, separation of enzymes from products, and preventing contamination. This process enables multi-enzyme systems for biosensors. The choice of immobilization method in biosensors depends on factors like the biological element, transducer type, analyte properties, and operating conditions [165]. Efficient enzyme immobilization offers benefits like reusing enzymes, improving stability, easy reaction control, separation of enzymes from products, and preventing contamination. This process enables multi-enzyme systems for biosensors. The choice of immobilization method in biosensors depends on factors like the biological element, transducer type, analyte properties, and operating conditions [165]. As a result, the choice of the most commonly used method for immobilizing laccase onto nanoparticles in water treatment applications varied based on specific research objectives and the nature of the contaminants being treated. However, covalent binding and encapsulation methods have been gaining traction due to their advantages in stability and reusability. Encapsulation and covalent binding methods have been explored for the immobilization of enzymes for the removal of micropollutants from water and wastewater. Encapsulation involves entrapping the enzyme within a support material, while covalent binding involves forming a chemical bond between the enzyme and the support material. Both methods have their advantages and challenges, and their favorability may depend on specific applications and the properties of the enzymes and support materials being used. Further research and development are needed to determine which method is more favorable for different scenarios and to improve the efficiency and practical application of enzyme-based technologies for wastewater treatment [166]. Covalent binding has gained popularity because of its robustness in preventing enzyme leaching and providing enhanced stability to the immobilized laccase [167]. Encapsulation, on the other hand, protects the enzyme within a matrix, shielding it from harsh environmental conditions. This method has been explored due to its potential to improve enzyme stability, particularly when dealing with contaminants in challenging conditions [168].

5. Applications of Immobilized Laccase in water treatment

Laccase immobilization presents considerable promise across diverse sectors. Notably, laccase-immobilized particles have showcased remarkable efficacy in the environmental and food industries. Within the textile sector, immobilized laccase finds utility in dye decolorization, providing an eco-friendly and cost-effective remedy. Beyond these domains, the applications of laccase immobilization have extended into pharmaceuticals, agriculture, baking, water treatment, and more. While the enhanced stability of immobilized laccases is a general advantage, challenges and prospects remain areas of exploration [179]. Laccase, when immobilized, has emerged as a highly effective tool in various applications, particularly in water treatment processes. The immobilization of laccase offers several advantages, such as enhanced stability, recyclability, and improved enzymatic activity, which make it a valuable asset for addressing water pollution challenges. Here are some notable applications of immobilized laccase in water treatment: Removal of organic contaminants, removal of azo dyes, removal of heavy metals

Immobilized laccase has shown promising results in the removal of organic pollutants from water treatment. It effectively degrades a wide range of compounds including pharmaceuticals, pesticides, dyes, and phenolic compounds. The enzymatic activity of immobilized laccase helps in breaking down these contaminants, leading to their degradation into less harmful substances [180].

Azo dyes, commonly used in textile and dye industries, pose a significant environmental concern due to their toxicity and resistance to conventional treatment methods. Immobilized laccase has demonstrated excellent potential in the degradation of azo dyes, showing remarkable efficiency in breaking down complex chemical structures and facilitating their removal from wastewater [181, 182].

Heavy metal contamination is a pressing issue in water bodies, and their presence poses severe health risks. Immobilized laccase, in combination with suitable chelating agents, can effectively remove heavy metals by facilitating their precipitation or transformation into less toxic forms. This approach offers a sustainable and eco-friendly alternative to conventional metal removal techniques [183].

M. Mazur et al. [184] in their finding introduced a simple method to firmly attach laccase to various conductive surfaces, preserving its enzyme activity. This relies on ionic coordination between zirconium phosphonate and protein carboxylate groups. Using multiple techniques, including SPR, QCM gravimetry, AFM, SERS, RR, and electrochemistry, we've shown that laccase, when linked to these surfaces via ZPC interactions, forms a stable enzymatic layer, maintaining its activity. However, this surface attachment seems to alter its bioactivity, leading to higher Km values in enzyme kinetics data. This change is a known outcome of the formation of a diffusion layer on solid surfaces. Consequently, the observed rise in Km values for laccase on ITO or Au surfaces can be attributed to enzyme conformational shifts or increased diffusion constraints.

Sanjay K. S. Patel et al. [185] and Qingqing Wang et al. [38] Noted the growing interest in employing metal-based supports for laccase immobilization. Describes the adsorption of metal-chelated enzymes, which relies on interactions between metal ions present on the support and specific enzyme groups. Diverse metal-based supports including Cu, Ca, Al, and Zr variants have been investigated for laccase immobilization, demonstrating enhancements in laccase activity and stability.

Chengyu Zhang et al. [81] mentioned that carbon-based materials, such as porous carbon materials, carbon nanotubes, and graphene, have been widely used as carriers for enzyme immobilization. These carbon materials offer excellent chemical stability, good adsorption capacity, and high conductivity, making them suitable for applications in enzyme immobilization and catalysis. Advantages of laccase immobilization: The document states that the immobilization of laccase on carbon-based carriers, such as magnetic carriers, can help overcome the limitations of free laccase, such as easy denaturation, poor stability, and difficulty in reuse. Immobilized laccase offers advantages such as improved thermal stability, higher productivity, cheaper cost, and easier purification, making it suitable for industrial production. Sanjay K. S. Patel et al. [21] showed Laccase immobilization on Cu/FeO4 NPs improved enzyme activity. Combining free R. vernicifera laccase with Cu (5 mM) resulted in a 1.5-fold activity increase. Magnetic NP-immobilized enzymes could be easily separated using a magnetic field. Immobilization methods involved Cu and Cu/FeO4 NPs, functionalized with glutaraldehyde, APTES, or APTES followed by glutaraldehyde. Optimal conditions were found at pH 5.0, achieving an impressive 93.1% immobilization yield (IY) and 140% relative activity (RA). This boost in RA was linked to the Cu metal ions' presence. In contrast, other methods like chitosan-based support and nylon membranes had lower IY and RA values. Temperature and incubation time played a role. An incubation temperature of 4–16°C maintained high IY (93.1–93.8%), but RA varied. Longer incubation (up to 4 h) stabilized IY at 93.3%. Cu/FeO4 NPs outperformed other methods, allowing for a maximum laccase immobilization of 85 mg/g of support at a loading rate of 600 mg of protein/g of support, attributed to smaller NP size. Impressively, at maximum loading, laccase exhibited a 105% RA compared to its free form, surpassing other reported laccase immobilization methods.

Rui Zhai et al. [186] focused on developing a modified poly(vinylidene fluoride) membrane (PVDF) with strong chemical stability and mechanical strength for laccase immobilization through covalent bonding. This is achieved by grafting 3-triethoxysilylpropylamine (APTES) modified FeO_3@SiO cubes (FS@cubes) onto the PDA layer using a solvothermal process, resulting in the formation of FS@cubes-PDA@PVDF membrane. Laccase is then immobilized on this surface via glutaraldehyde (GA) crosslinking, resulting in Lac-FS@cubes-PDA@PVDF. Under optimal conditions (pH 7.0 and 35 ℃), Lac-FS@cubes-PDA@PVDF demonstrates a high removal efficiency of 97.1% for the pollutant Congo red, surpassing the performance of free laccase. Furthermore, this immobilized system exhibits excellent stability even after low-temperature storage and can be reused effectively. This finding suggests a potential strategy for removing various water pollutants and provides a straightforward approach for large-scale applications of enzyme-catalysis in water treatment.

Nerea Ormategui et al. [187] focused on creating stable and efficient nano biocatalysts using laccase enzyme immobilized on composite hydrogels consisting of (rGO) and a polymer matrix. The composite hydrogel supports were synthesized through the self-assembly of (GO) nanoplatelets within a polymer latex matrix, forming hybrid nanoplatelets. Ascorbic acid was used as a reducing agent for (GO), resulting in the formation of three-dimensional porous structures - the composite hydrogels. These hydrogels served as a support for covalently immobilizing laccase. The performance of these nano biocatalysts was evaluated in the oxidative degradation of a stubborn synthetic dye called Remazol Brilliant Blue R in water. The biocatalysts exhibited significant dye discoloration capabilities and remained highly stable, retaining their catalytic activity across four successive batches of dye degradation. This approach presents a promising solution to address the common challenges associated with enzyme catalysts, including their fragility, cost, and the need for high enzyme loading, thereby advancing their potential for industrial applications. Yannick-Serge Zimmermann et al. [188] explore a method to enhance the performance of laccase enzymes in eliminating micropollutants from wastewater. They achieve this by immobilizing laccase enzymes from Coriolopsis polyzona onto solid surfaces, specifically amino-modified silica nanoparticles, using glutaraldehyde for cross-linking. The results of this method are highly promising. The immobilized laccase exhibits remarkable stability, with 77% of its activity retained in real wastewater over a month. In contrast, free laccase, without immobilization, retained only .5% of its activity under the same conditions. The success of this novel approach suggests its potential for efficient micropollutant removal in wastewater treatment, emphasizing the significance of enzyme immobilization in environmental applications

Runtang Liu et al. [161] in their article discuss the use of magnetic nanoparticles in the immobilization of laccase, an enzyme used for the removal of phenolic pollutants. The study focuses on combining magnetic Fe_3O_4 nanoparticles with polyethyleneimine (PEI) through the bridging of carboxyl-functionalized ionic liquid. The resulting magnetic polyethyleneimine nanoparticles (MPEI) show good immobilization ability, with laccase loading and activity retention reaching 5.19 mg/g and 91.65%, respectively. The use of magnetic nanoparticles provides advantages such as easy separation, good mechanical stability, and low toxicity, making them suitable for laccase immobilization.

Raquel A. Fernandes et al. [189] Introduced novel magnetic nanoparticles (MNPs) that were synthesized via EDTA-TMS functionalization and comprehensively characterized using techniques like TEM, FTIR, and BET analysis. These MNPs were utilized as a support matrix for laccase immobilization, showcasing a promising development in biocatalysis. Despite EDTA-TMS's known chelating properties, its use for modifying MNPs to immobilize laccase is a novel strategy, presented here for the first time. The immobilization process exhibited around 97% recovery of enzymatic activity at pH 3.5. The immobilized laccase displayed altered Michaelis-Menten kinetics, with a lower Vmax and similar KM compared to its free form. In terms of stability, the immobilized enzyme retained approximately 73% of its initial activity after five consecutive reaction cycles. Furthermore, this immobilized enzyme efficiently catalyzed the degradation of Indigo Carmine dye. These MNPs hosting immobilized laccase demonstrated notable advantages over other materials, highlighting their potential applications in industrial biochemical processes, biocatalysis, and biosensors.

Another research by María Fernández-Fernández et al. [190] reported metal-based supports such as gold, silver, indium tin oxide, zirconium-phosphonate-carboxylate, and mesoporous silica. These metal-based supports were functionalized and used to immobilize laccase, resulting in improved pH and thermal stability of the enzyme. The immobilized laccase showed potential for applications such as enzymatic reduction of dioxygen, degradation of hydroxylated compounds, and decolorization of dyes. The article mentions different methods for immobilizing laccase, including coordination chemistry, self-assembled monolayers, sol-gel silica, layered double hydroxides, and thermoresponsive gels. These methods involve the use of various supports and functionalization techniques to immobilize laccase, resulting in improved stability and activity of the enzyme.

6. Conclusions and future insights

The field of nanoparticle-supported laccase immobilization presents a landscape of opportunities and innovations. This technique has demonstrated its significance, particularly in the context of water treatment, but its potential reaches far beyond that Immobilizing laccase on nanoparticles enhances its capabilities, making it a robust and versatile tool for various applications like water treatment, pharmaceuticals, agriculture, and food industry. Nanoparticle Immobilization methods employed for nanoparticle-supported laccase immobilization contribute to its success. Physical adsorption, covalent binding, encapsulation, cross-linking, and layer-by-layer assembly provide versatile choices. The advantages here are flexibility and adaptability, allowing tailoring of the immobilization process to specific needs and contexts. The selection of nanoparticles, including metal-based, magnetic, carbon-based, and others, is crucial. Each type brings unique advantages, such as enhanced stability and catalytic activity. Metal-based supports, for example, exhibit improved laccase activity and stability, while carbon-based materials offer excellent chemical stability and high conductivity, making them ideal for enzyme immobilization looking ahead, the future of nanoparticle-supported laccase immobilization is filled with exciting prospects. Researchers and scientists are encouraged to explore and refine the techniques involved. Further optimization of immobilization methods, the investigation of new nanoparticles, and the development of innovative surface modifications can push the boundaries of enzyme activity and stability. The extensive benefits of laccase immobilization supported by nanoparticles extend to multiple sectors. Its capacity to improve efficiency, reduce environmental impact, and offer economic advantages makes it a promising and versatile solution. As research in this field progresses, we can anticipate further refinements and applications that will continue to shape a more sustainable and eco-conscious future.

Acknowledgments

The authors received no financial support for the research, author-ship and/or publication of this article.

Conflict of interest

The authors declare that there is no conflict of interest.

REFERENCES

[1] M. Alcalde, Laccases: biological functions, molecular structure and industrial applications, Industrial Enzymes: structure, function and applications, Springer 2007, pp. 461-476.
[2] S.F.F. Zofair, S. Ahmad, M.A. Hashmi, S.H. Khan, M.A. Khan, H. Younus, Catalytic roles, immobilization and management of recalcitrant environmental pollutants by laccases: significance in sustainable green chemistry, Journal of Environmental Management 309 (2022) 114676.
[3] W. Zhou, W. Zhang, Y. Cai, Laccase immobilization for water purification: A comprehensive review, Chemical Engineering Journal 403 (2021) 126272.
[4] K. Thakur, C. Attri, A. Seth, Nanocarriers-based immobilization of enzymes for industrial application, 3 Biotech 11(10) (2021) 427.
[5] D. Ren, Z. Wang, S. Jiang, H. Yu, S. Zhang, X. Zhang, Recent environmental applications of and development prospects for immobilized laccase: a review, Biotechnology and Genetic Engineering Reviews 36(2) (2020) 81-131.
[6] S.M. Dizaj, A. Mennati, S. Jafari, K. Khezri, K. Adibkia, Antimicrobial activity of carbon-based nanoparticles, Advanced pharmaceutical bulletin 5(1) (2015) 19.
[7] L. Najmi, Z. Hu, Review on Molecular Dynamics Simulations of Effects of Carbon Nanotubes (CNTs) on Electrical and Thermal Conductivities of CNT-Modified Polymeric Composites, Journal of Composites Science 7(4) (2023) 165.
[8] M. Azizi-Lalabadi, H. Hashemi, J. Feng, S.M. Jafari, Carbon nanomaterials against pathogens; the antimicrobial activity of carbon nanotubes, graphene/graphene oxide, fullerenes, and their nanocomposites, Advances in Colloid and Interface Science 284 (2020) 102250.

[9] A. Ali, T. Shah, R. Ullah, P. Zhou, M. Guo, M. Ovais, Z. Tan, Y. Rui, Review on recent progress in magnetic nanoparticles: Synthesis, characterization, and diverse applications, Frontiers in Chemistry 9 (2021) 629054.
[10] N.A. Daronch, M. Kelbert, C.S. Pereira, P.H.H. de Araújo, D. de Oliveira, Elucidating the choice for a precise matrix for laccase immobilization: A review, Chemical Engineering Journal 397 (2020) 125506.
[11] P. Singh, A. Borthakur, A review on biodegradation and photocatalytic degradation of organic pollutants: A bibliometric and comparative analysis, Journal of cleaner production 196 (2018) 1669-1680.
[12] F. Bialas, D. Reichinger, C.F. Becker, Biomimetic and biopolymer-based enzyme encapsulation, Enzyme and Microbial Technology 150 (2021) 109864.
[13] M. Ajith, M. Aswathi, E. Priyadarshini, P. Rajamani, Recent innovations of nanotechnology in water treatment: A comprehensive review, Bioresource Technology 342 (2021) 126000.
[14] T.M. Joseph, H.E. Al-Hazmi, B. Śniatała, A. Esmaeili, S. Habibzadeh, Nanoparticles and nanofiltration for wastewater treatment: From polluted to fresh water, Environmental Research (2023) 117114.
[15] Q. Zhu, J. Song, Z. Liu, K. Wu, X. Li, Z. Chen, H. Pang, Photothermal catalytic degradation of textile dyes by laccase immobilized on Fe_3O_4@SiO_2 nanoparticles, Journal of Colloid and Interface Science 623 (2022) 992-1001.
[16] Z. Chen, J. Yao, B. Ma, B. Liu, J. Kim, H. Li, X. Zhu, C. Zhao, M. Amde, A robust biocatalyst based on laccase immobilized superparamagnetic Fe3O4@ SiO_2–NH_2 nanoparticles and its application for degradation of chlorophenols, Chemosphere 291 (2022) 132727.
[17] Y. Gao, M. Wang, K. Shah, S. Singh Kalra, L.H. Rome, S. Mahendra, Decolorization and detoxification of synthetic dye compounds by laccase immobilized in vault nanoparticles, Bioresource Technology 351 (2022) 127040.
[18] Z. Chen, W.-D. Oh, P.-S. Yap, Recent advances in the utilization of immobilized laccase for the degradation of phenolic compounds in aqueous solutions: A review, Chemosphere 307 (2022) 135824.
[19] M. Iqhrammullah, A. Fahrina, W. Chiari, K. Ahmad, F. Fitriani, N. Suriaini, E. Safitri, K. Puspita, Laccase Immobilization Using Polymeric Supports for Wastewater Treatment: A Critical Review, Macromolecular Chemistry and Physics 224(9) (2023) 2200461.
[20] Z. Han, X. Fan, S. Yu, X. Li, S. Wang, L. Lu, Metal-organic frameworks (MOFs): A novel platform for laccase immobilization and application, Journal of Environmental Chemical Engineering 10(6) (2022) 108795.
[21] S.K.S. Patel, V.C. Kalia, J.K. Lee, Laccase Immobilization on Copper-Magnetic Nanoparticles for Efficient Bisphenol Degradation, J Microbiol Biotechnol 33(1) (2023) 127-134.
[22] D. Pandey, A. Daverey, K. Dutta, K. Arunachalam, Bioremoval of toxic malachite green from water through simultaneous decolorization and degradation using laccase immobilized biochar, Chemosphere 297 (2022) 134126.
[23] Y. Feng, Y. Xu, S. Liu, D. Wu, Z. Su, G. Chen, J. Liu, G. Li, Recent advances in enzyme immobilization based on novel porous framework materials and its applications in biosensing, Coordination Chemistry Reviews 459 (2022) 214414.
[24] S. Liang, X.-L. Wu, J. Xiong, M.-H. Zong, W.-Y. Lou, Metal-organic frameworks as novel matrices for efficient enzyme immobilization: an update review, Coordination Chemistry Reviews 406 (2020) 213149.
[25] A. Sirelkhatim, S. Mahmud, A. Seeni, N.H.M. Kaus, L.C. Ann, S.K.M. Bakhori, H. Hasan, D. Mohamad, Review on Zinc Oxide Nanoparticles: Antibacterial Activity and Toxicity Mechanism, Nano-Micro Letters 7(3) (2015) 219-242.
[26] T. Parandhaman, M.D. Dey, S.K. Das, Biofabrication of supported metal nanoparticles: exploring the bioinspiration strategy to mitigate the environmental challenges, Green Chemistry 21(20) (2019) 5469-5500.
[27] E. Sánchez-López, D. Gomes, G. Esteruelas, L. Bonilla, A.L. Lopez-Machado, R. Galindo, A. Cano, M. Espina, M. Ettcheto, A. Camins, A.M. Silva, A. Durazzo, A. Santini, M.L. Garcia, E.B. Souto, Metal-Based Nanoparticles as Antimicrobial Agents: An Overview, Nanomaterials 10(2) (2020) 292.
[28] H.D. Kyomuhimbo, H.G. Brink, Applications and immobilization strategies of the copper-centred laccase enzyme; a review, Heliyon (2023).
[29] P. Kumar, A. Deep, K.-H. Kim, Metal organic frameworks for sensing applications, TrAC Trends in Analytical Chemistry 73 (2015) 39-53.
[30] C. Wang, J. Luan, C. Wu, Metal-organic frameworks for aquatic arsenic removal, Water research 158 (2019) 370-382.
[31] Z. Yin, S. Wan, J. Yang, M. Kurmoo, M.-H. Zeng, Recent advances in post-synthetic modification of metal–organic frameworks: New types and tandem reactions, Coordination Chemistry Reviews 378 (2019) 500-512.
[32] M. Hua, S. Zhang, B. Pan, W. Zhang, L. Lv, Q. Zhang, Heavy metal removal from water/wastewater by nanosized metal oxides: A review, Journal of Hazardous Materials 211-212 (2012) 317-331.
[33] K. Gupta, P. Joshi, R. Gusain, O.P. Khatri, Recent advances in adsorptive removal of heavy metal and metalloid ions by metal oxide-based nanomaterials, Coordination Chemistry Reviews 445 (2021) 214100.
[34] S. Ren, F. Wang, H. Gao, X. Han, T. Zhang, Y. Yuan, Z. Zhou, Recent progress and future prospects of laccase immobilization on MOF supports for industrial applications, Applied Biochemistry and Biotechnology (2023) 1-16.
[35] M. Bilal, S.S. Ashraf, J. Cui, W.-Y. Lou, M. Franco, S.I. Mulla, H.M. Iqbal, Harnessing the biocatalytic attributes and applied perspectives of nanoengineered laccases—A review, International Journal of Biological Macromolecules 166 (2021) 352-373.
[36] L. Lei, X. Yang, Y. Song, H. Huang, Y. Li, Current research progress on laccase-like nanomaterials, New Journal of Chemistry 46(8) (2022) 3541-3550.
[37] T.-Q. Chai, J.-L. Wang, G.-Y. Chen, L.-X. Chen, F.-Q. Yang, Tris-Copper Nanozyme as a Novel Laccase Mimic for the Detection and Degradation of Phenolic Compounds, Sensors 23(19) (2023) 8137.
[38] Q. Wang, J. Cui, G. Li, J. Zhang, F. Huang, Q. Wei, Laccase immobilization by chelated metal ion coordination chemistry, Polymers 6(9) (2014) 2357-2370.
[39] D. Li, Y. Cheng, H. Zuo, W. Zhang, G. Pan, Y. Fu, Q. Wei, Dual-functional biocatalytic membrane containing laccase-embedded metal-organic frameworks for detection and degradation of phenolic pollutant, Journal of Colloid and Interface Science 603 (2021) 771-782.
[40] C. Carnovale, G. Bryant, R. Shukla, V. Bansal, Size, shape and surface chemistry of nano-gold dictate its cellular interactions, uptake and toxicity, Progress in Materials Science 83 (2016) 152-190.
[41] M. Drozd, A. Duszczyk, P. Ivanova, M. Pietrzak, Interactions of proteins with metal-based nanoparticles from a point of view of analytical chemistry-Challenges and opportunities, Advances in Colloid and Interface Science 304 (2022) 102656.
[42] F. Ahmad, M.M. Salem-Bekhit, F. Khan, S. Alshehri, A. Khan, M.M. Ghoneim, H.-F. Wu, E.I. Taha, I. Elbagory, Unique properties of surface-functionalized nanoparticles for bio-application: Functionalization mechanisms and importance in application, Nanomaterials 12(8) (2022) 1333.
[43] C.C.S. Fortes, A.L. Daniel-da-Silva, A.M.R.B. Xavier, A.P.M. Tavares, Optimization of enzyme immobilization on functionalized magnetic nanoparticles for laccase biocatalytic reactions, Chemical Engineering and Processing: Process Intensification 117 (2017) 1-8.
[44] A. Ali, T. Shah, R. Ullah, P. Zhou, M. Guo, M. Ovais, Z. Tan, Y. Rui, Review on Recent Progress in Magnetic Nanoparticles: Synthesis, Characterization, and Diverse Applications, Frontiers in Chemistry 9 (2021).
[45] M.A. Mariño, S. Fulaz, L. Tasic, Magnetic Nanomaterials as Biocatalyst Carriers for Biomass Processing: Immobilization Strategies, Reusability, and Applications, Magnetochemistry 7(10) (2021) 133.
[46] Y. Liu, Z. Zeng, G. Zeng, L. Tang, Y. Pang, Z. Li, C. Liu, X. Lei, M. Wu, P. Ren, Z. Liu, M. Chen, G. Xie, Immobilization of laccase on magnetic bimodal mesoporous carbon and the application in the removal of phenolic compounds, Bioresource Technology 115 (2012) 21-26.
[47] H. Wu, W. Mu, Application prospects and opportunities of inorganic nanomaterials for enzyme immobilization in the food-processing industry, Current Opinion in Food Science 47 (2022) 100909.
[48] S. Cheriyamundath, S.L. Vavilala, Nanotechnology-based wastewater treatment, Water and Environment Journal 35(1) (2021) 123-132.
[49] S. Aziz, A. Abdel-Karim, Dual-functional ultrafiltration biocatalytic membrane containing laccase/ nanoparticle for removal of pollutants: A review, Environmental Nanotechnology, Monitoring & Management 20 (2023) 100852.
[50] L. Xie, R. Jiang, F. Zhu, H. Liu, G. Ouyang, Application of functionalized magnetic nanoparticles in sample preparation, Analytical and Bioanalytical Chemistry 406(2) (2014) 377-399.
[51] S. Sharma, A. Verma, A. Kumar, H. Kamyab, Magnetic Nano-Composites and their Industrial Applications, Nano Hybrids and Composites 20 (2018) 149-172.
[52] P.A. Johnson, H.J. Park, A.J. Driscoll, Enzyme nanoparticle fabrication: magnetic nanoparticle synthesis and enzyme immobilization, Enzyme Stabilization and Immobilization: Methods and Protocols (2011) 183-191
[53] C. Ottone, O. Romero, C. Aburto, A. Illanes, L. Wilson, Biocatalysis in the winemaking industry: Challenges and opportunities for immobilized enzymes, Comprehensive reviews in food science and food safety 19(2) (2020) 595-621.
[54] S.K.S. Patel, R.K. Gupta, S.-Y. Kim, I.-W. Kim, V.C. Kalia, J.-K. Lee, Rhus vernicifera Laccase Immobilization on Magnetic Nanoparticles to Improve Stability and Its Potential Application in Bisphenol A Degradation, Indian Journal of Microbiology 61(1) (2021) 45-54.
[55] S. Mojtabavi, F. Rezayaraghi, M. Shahverdi, H.T. Golroudbari, F. Moshiri, M.A. Faramarzi, Bioremoval and biodetoxification of ciprofloxacin from hospital wastewater effluent by the efficient and recyclable laccase@ hercynite magnetic nanoparticles, Journal of Environmental Chemical Engineering (2023) 111599.
[56] D.C. Sotelo, N. Ornelas-Soto, J.F. Osma, Novel Magnetic Polymeric Filters

with Laccase-Based Nanoparticles for Improving Congo Red Decolorization in Bioreactors, Polymers 14(12) (2022) 2328.
[57] A. Kumari, R. Rajeev, L. Benny, Y. Sudhakar, A. Varghese, G. Hegde, Recent advances in carbon nanotubes-based biocatalysts and their applications, Advances in Colloid and Interface Science 297 (2021) 102542.
[58] S.N. Patel, V. Singh, M. Sharma, R.S. Sangwan, N.K. Singhal, S.P. Singh, Development of a thermo-stable and recyclable magnetic nanobiocatalyst for bioprocessing of fruit processing residues and D-allulose synthesis, Bioresource technology 247 (2018) 633-639.
[59] K. Meller, M. Szumski, B. Buszewski, Microfluidic reactors with immobilized enzymes—Characterization, dividing, perspectives, Sensors and actuators B: Chemical 244 (2017) 84-106.
[60] B. Bayatsarmadi, Y. Zheng, A. Vasileff, S.-Z. Qiao, Recent Advances in Atomic Metal Doping of Carbon-based Nanomaterials for Energy Conversion, Small 13(21) (2017) 1700191.
[61] Q. Wu, L. Yang, X. Wang, Z. Hu, From Carbon-Based Nanotubes to Nanocages for Advanced Energy Conversion and Storage, Accounts of Chemical Research 50(2) (2017) 435-444.
[62] F. Ameen, K. Alsamhary, J.A. Alabdullatif, S. Alnadhari, A review on metal-based nanoparticles and their toxicity to beneficial soil bacteria and fungi, Ecotoxicology and Environmental Safety 213 (2021) 112027.
[63] B.-T. Zhang, X. Zheng, H.-F. Li, J.-M. Lin, Application of carbon-based nanomaterials in sample preparation: A review, Analytica Chimica Acta 784 (2013) 1-17.
[64] M.S. Mauter, M. Elimelech, Environmental Applications of Carbon-Based Nanomaterials, Environmental Science & Technology 42(16) (2008) 5843-5859.
[65] C.J. Shearer, A. Cherevan, D. Eder, Application and Future Challenges of Functional Nanocarbon Hybrids, Advanced Materials 26(15) (2014) 2295-2318.
[66] K. Scida, P.W. Stege, G. Haby, G.A. Messina, C.D. García, Recent applications of carbon-based nanomaterials in analytical chemistry: Critical review, Analytica Chimica Acta 691(1) (2011) 6-17.
[67] X. Gong, Y. Meng, J. Lu, Y. Tao, Y. Cheng, H. Wang, A Review on lignin-based phenolic resin adhesive, Macromolecular Chemistry and Physics 223(4) (2022) 2100434.
[68] L.Y. Ng, A.W. Mohammad, C.P. Leo, N. Hilal, Polymeric membranes incorporated with metal/metal oxide nanoparticles: A comprehensive review, Desalination 308 (2013) 15-33.
[69] K. Gong, Y. Yan, M. Zhang, L. Su, S. Xiong, L. Mao, Electrochemistry and Electroanalytical Applications of Carbon Nanotubes: A Review, Analytical Sciences 21(12) (2005) 1383-1393.
[70] S. Cheriyamundath, S.L. Vavilala, Nanotechnology-based wastewater treatment, Water and Environment Journal 35(1) (2021) 123-132.
[71] S.S. Fiyadh, M.A. AlSaadi, W.Z. Jaafar, M.K. AlOmar, S.S. Fayaed, N.S. Mohd, L.S. Hin, A. El-Shafie, Review on heavy metal adsorption processes by carbon nanotubes, Journal of Cleaner Production 230 (2019) 783-793.
[72] X. Yang, Y. Wan, Y. Zheng, F. He, Z. Yu, J. Huang, H. Wang, Y.S. Ok, Y. Jiang, B. Gao, Surface functional groups of carbon-based adsorbents and their roles in the removal of heavy metals from aqueous solutions: A critical review, Chemical Engineering Journal 366 (2019) 608-621.
[73] M.S. Soffian, F.Z. Abdul Halim, F. Aziz, M. A. Rahman, M.A. Mohamed Amin, D.N. Awang Chee, Carbon-based material derived from biomass waste for wastewater treatment, Environmental Advances 9 (2022) 100259.
[74] K.P. Gopinath, D.-V.N. Vo, D. Gnana Prakash, A. Adithya Joseph, S. Viswanathan, J. Arun, Environmental applications of carbon-based materials: a review, Environmental chemistry letters 19 (2021) 557-582.
[75] N. Madima, S. Mishra, I. Inamuddin, A. Mishra, Carbon-based nanomaterials for remediation of organic and inorganic pollutants from wastewater. A review, Environmental Chemistry Letters 18 (2020) 1169-1191.s
[76] I.V. Pavlidis, T. Vorhaben, T. Tsoufis, P. Rudolf, U.T. Bornscheuer, D. Gournis, H. Stamatis, Development of effective nanobiocatalytic systems through the immobilization of hydrolases on functionalized carbon-based nanomaterials, Bioresource Technology 115 (2012) 164-171.
[77] K. Scida, P.W. Stege, G. Haby, G.A. Messina, C.D. García, Recent applications of carbon-based nanomaterials in analytical chemistry: critical review, Analytica Chimica Acta 691(1-2) (2011) 6-17.
[78] Y. Adamian, L. Lonappan, K. Alokpa, S.N. Agathos, H. Cabana, Recent Developments in the Immobilization of Laccase on Carbonaceous Supports for Environmental Applications - A Critical Review, Frontiers in Bioengineering and Biotechnology 9 (2021).
[79] J. Wu, F. Xu, S. Li, P. Ma, X. Zhang, Q. Liu, R. Fu, D. Wu, Porous Polymers as Multifunctional Material Platforms toward Task-Specific Applications, Advanced Materials 31(4) (2019) 1802922.
[80] A.A. Kadam, G.D. Saratale, G.S. Ghodake, R.G. Saratale, A. Shahzad, V.K. Magotra, M. Kumar, R.R. Palem, J.-S. Sung, Recent Advances in the Development of Laccase-Based Biosensors via Nano-Immobilization Techniques, Chemosensors 10(2) (2022) 58.
[81] C. Zhang, S. You, Y. Liu, C. Wang, Q. Yan, W. Qi, R. Su, Z. He, Construction of luffa sponge-based magnetic carbon nanocarriers for laccase immobilization and its application in the removal of bisphenol A, Bioresource Technology 305 (2020) 123085.
[82] M. Patila, P.E. Athanasiou, L. Kortessis, G. Potsi, A. Kouloumpis, D. Gournis, H. Stamatis, Immobilization of Laccase on Hybrid Super-Structured Nanomaterials for the Decolorization of Phenolic Dyes, Processes 10(2) (2022) 233.
[83] L. Najmi, S.M. Zebarjad, K. Janghorban, Effects of Carbon Nanotubes on the Compressive and Flexural Strength and Microscopic Structure of Epoxy Honeycomb Sandwich Panels, Polymer Science, Series B (2023) 1-10.
[84] R. Sangubotla, J. Kim, Fiber-optic biosensor based on the laccase immobilization on silica-functionalized fluorescent carbon dots for the detection of dopamine and multi-color imaging applications in neuroblastoma cells, Materials Science and Engineering: C 122 (2021) 111916.
[85] M. Masjoudi, M. Golgoli, Z.G. Nejad, S. Sadeghzadeh, S.M. Borghei, Pharmaceuticals removal by immobilized laccase on polyvinylidene fluoride nanocomposite with multi-walled carbon nanotubes, Chemosphere 263 (2021) 128043.
[86] W. Zhang, Q. Yang, Q. Luo, L. Shi, S. Meng, Laccase-Carbon nanotube nanocomposites for enhancing dyes removal, Journal of Cleaner Production 242 (2020) 118425.
[87] M.L. Verma, Sukriti, B. Dhanya, R. Saini, A. Das, R.S. Varma, Synthesis and application of graphene-based sensors in biology: a review, Environmental Chemistry Letters 20(3) (2022) 2189-2212.
[88] Y. Adamian, L. Lonappan, K. Alokpa, S.N. Agathos, H. Cabana, Recent developments in the immobilization of laccase on carbonaceous supports for environmental applications-a critical review, Frontiers in Bioengineering and Biotechnology 9 (2021) 778239.
[89] V. Califano, A. Costantini, Immobilization of cellulolytic enzymes in mesostructured silica materials, Catalysts 10(6) (2020) 706.
[90] S. Aggarwal, A. Chakravarty, S. Ikram, A comprehensive review on incredible renewable carriers as promising platforms for enzyme immobilization & thereof strategies, International Journal of Biological Macromolecules 167 (2021) 962-986.
[91] R. Yadav, T. Baskaran, A. Kaiprathu, M. Ahmed, S.V. Bhosale, S. Joseph, A.a.H. Al-Muhtaseb, G. Singh, A. Sakthivel, A. Vinu, Recent advances in the preparation and applications of organo-functionalized porous materials, Chemistry–An Asian Journal 15(17) (2020) 2588-2621.
[92] Y. Hui, Z. Huang, M.E.E. Alahi, A. Nag, S. Feng, S.C. Mukhopadhyay, Recent advancements in electrochemical biosensors for monitoring the water quality, Biosensors 12(7) (2022) 551.
[93] K.A. Al-Maqdi, N. Elmerhi, K. Athamneh, M. Bilal, A. Alzamly, S.S. Ashraf, I. Shah, Challenges and recent advances in enzyme-mediated wastewater remediation—a review, Nanomaterials 11(11) (2021) 3124.
[94] M. Mozetič, A. Vesel, G. Primc, C. Eisenmenger-Sittner, J. Bauer, A. Eder, G.H. Schmid, D.N. Ruzic, Z. Ahmed, D. Barker, Recent developments in surface science and engineering, thin films, nanoscience, biomaterials, plasma science, and vacuum technology, Thin Solid Films 660 (2018) 120-160.
[95] A. Zielińska, F. Carreiró, A.M. Oliveira, A. Neves, B. Pires, D.N. Venkatesh, A. Durazzo, M. Lucarini, P. Eder, A.M. Silva, A. Santini, E.B. Souto, Polymeric Nanoparticles: Production, Characterization, Toxicology and Ecotoxicology, Molecules 25(16) (2020) 3731.
[96] O. Dumbrava, A. Filimon, L. Marin, Tailoring properties and applications of polysulfone membranes by chemical modification: Structure-properties-applications relationship, European Polymer Journal (2023) 112316.
[97] S. Nosheen, M. Irfan, S.H. Abidi, Q. Syed, F. Habib, A. Asghar, B. Waseem, B. Soomro, H. Butt, M. Akram, A review: Development of magnetic nano vectors for biomedical applications, GSC Advanced Research and Reviews 8(2) (2021) 085-110.
[98] M. Li, Q. Shi, N. Song, Y. Xiao, L. Wang, Z. Chen, T.D. James, Current trends in the detection and removal of heavy metal ions using functional materials, Chemical Society Reviews (2023).
[99] M. Qamar, A. Basharat, S.A. Qamar, M. Bilal, M. Franco, H.M.N. Iqbal, Enzyme-loaded nanostructured materials for the degradation of environmental pollutants, Current Opinion in Environmental Science & Health 30 (2022) 100400.
[100] Y. Zhu, F. Qiu, J. Rong, T. Zhang, K. Mao, D. Yang, Covalent laccase immobilization on the surface of poly (vinylidene fluoride) polymer membrane for enhanced biocatalytic removal of dyes pollutants from aqueous environment, Colloids and Surfaces B: Biointerfaces 191 (2020) 111025.

[101] M.A. Agotegaray, V.L. Lassalle, Silica-coated magnetic nanoparticles: an insight into targeted drug delivery and toxicology, Springer2017.
[102] A. Maio, I. Pibiri, M. Morreale, F.P.L. Mantia, R. Scaffaro, An overview of functionalized graphene nanomaterials for advanced applications, Nanomaterials 11(7) (2021) 1717.
[103] S. ul Haque, N. Duteanu, S. Ciocan, A. Nasar, A review: Evolution of enzymatic biofuel cells, Journal of Environmental Management 298 (2021) 113483.
[104] F. Wang, C. Guo, L.-r. Yang, C.-Z. Liu, Magnetic mesoporous silica nanoparticles: Fabrication and their laccase immobilization performance, Bioresource Technology 101(23) (2010) 8931-8935.
[105] J. Bebić, K. Banjanac, M. Ćorović, A. Milivojević, M. Simović, A. Marinković, D. Bezbradica, Immobilization of laccase from Myceliophthora thermophila on functionalized silica nanoparticles: Optimization and application in lindane degradation, Chinese Journal of Chemical Engineering 28(4) (2020) 1136-1144.
[106] S. Kashefi, S.M. Borghei, N.M. Mahmoodi, Covalently immobilized laccase onto graphene oxide nanosheets: Preparation, characterization, and biodegradation of azo dyes in colored wastewater, Journal of Molecular Liquids 276 (2019) 153-162.
[107] Q. Lei, J. Guo, A. Noureddine, A. Wang, S. Wuttke, C.J. Brinker, W. Zhu, Sol–gel-based advanced porous silica materials for biomedical applications, Advanced Functional Materials 30(41) (2020) 1909539.
[108] A. Popat, S.B. Hartono, F. Stahr, J. Liu, S.Z. Qiao, G.Q.M. Lu, Mesoporous silica nanoparticles for bioadsorption, enzyme immobilisation, and delivery carriers, Nanoscale 3(7) (2011) 2801-2818.
[109] R. Fopase, S. Paramasivam, P. Kale, B. Paramasivan, Strategies, challenges and opportunities of enzyme immobilization on porous silicon for biosensing applications, Journal of Environmental Chemical Engineering 8(5) (2020) 104266.
[110] J.C. Nunes, M.R. Almeida, R.M. Bento, M.M. Pereira, V.C. Santos-Ebinuma, M.C. Neves, M.G. Freire, A.P. Tavares, Enhanced enzyme reuse through the bioconjugation of L-asparaginase and silica-based supported ionic liquid-like phase materials, Molecules 27(3) (2022) 929.
[111] A. Nayl, A. Abd-Elhamid, A.A. Aly, S. Bräse, Recent progress in the applications of silica-based nanoparticles, RSC advances 12(22) (2022) 13706-13726.
[112] M.T. Amin, A.A. Alazba, U. Manzoor, A review of removal of pollutants from water/wastewater using different types of nanomaterials, Advances in materials science and engineering 2014 (2014) 1-24.
[113] K. Simeonidis, S. Mourdikoudis, E. Kaprara, M. Mitrakas, L. Polavarapu, Inorganic engineered nanoparticles in drinking water treatment: a critical review, Environmental Science: Water Research & Technology 2(1) (2016) 43-70.
[114] S.A. Younis, M.M. Ghobashy, G. Bassioni, A.K. Gupta, Tailored functionalized polymer nanoparticles using gamma radiation for selected adsorption of barium and strontium in oilfield wastewater, Arabian Journal of Chemistry 13(2) (2020) 3762-3774.
[115] X. Tan, X. Wang, Water treatment and environmental remediation applications of carbon-based nanomaterials, Emerging Nanomaterials for Recovery of Toxic and Radioactive Metal Ions from Environmental Media, Elsevier2022, pp. 229-311.
[116] R. Singh, M.S. Samuel, M. Ravikumar, S. Ethiraj, V.S. Kirankumar, M. Kumar, R. Arulvel, S. Suresh, Processing of Carbon-Based Nanomaterials for the Removal of Pollutants from Water/Wastewater Application, Water 15(16) (2023) 3003.
[117] S. Manimegalai, S. Vickram, S.R. Deena, K. Rohini, S. Thanigaivel, S. Manikandan, R. Subbaiya, N. Karmegam, W. Kim, M. Govarthanan, Carbon-based nanomaterial intervention and efficient removal of various contaminants from effluents–a review, Chemosphere 312 (2023) 137319.
[118] R. Das, S.B. Abd Hamid, M.E. Ali, A.F. Ismail, M. Annuar, S. Ramakrishna, Multifunctional carbon nanotubes in water treatment: the present, past and future, Desalination 354 (2014) 160-179.
[119] M. Mogharabi-Manzari, M. Kiani, S. Aryanejad, S. Imanparast, M. Amini, M.A. Faramarzi, A magnetic heterogeneous biocatalyst composed of immobilized laccase and 2, 2, 6, 6-tetramethylpiperidine-1-oxyl (TEMPO) for green one-pot cascade synthesis of 2-substituted benzimidazole and benzoxazole derivatives under mild reaction conditions, Advanced Synthesis & Catalysis 360(18) (2018) 3563-3571.
[120] N. Baig, I. Kammakakam, W. Falath, Nanomaterials: A review of synthesis methods, properties, recent progress, and challenges, Materials Advances 2(6) (2021) 1821-1871.
[121] Z. Farka, T. Jurik, D. Kovar, L. Trnkova, P. Skládal, Nanoparticle-based immunochemical biosensors and assays: recent advances and challenges, Chemical reviews 117(15) (2017) 9973-10042.
[122] T.A. Aragaw, A.A. Ayalew, Chapter 10 - Application of metal-based nanoparticles for metal removal for treatments of wastewater -- a review, in: A. Ahmad, R. Kumar, M. Jawaid (Eds.), Emerging Techniques for Treatment of Toxic Metals from Wastewater, Elsevier 2023, pp. 183-231.
[123] M. Rajamehala, A.M. Pandian, M. Rajasimman, B. Gopalakrishnan, Synthesis of metal-based functional nanocomposite material and its application for the elimination of paracetamol from synthetic wastewater, Chemosphere 308 (2022) 136530.
[124] H. An, M. Li, J. Gao, Z. Zhang, S. Ma, Y. Chen, Incorporation of biomolecules in Metal-Organic Frameworks for advanced applications, Coordination Chemistry Reviews 384 (2019) 90-106.
[125] F. Almomani, R. Bhosale, M. Khraisheh, A. kumar, T. Almomani, Heavy metal ions removal from industrial wastewater using magnetic nanoparticles (MNP), Applied Surface Science 506 (2020) 144924.
[126] S. Vallinayagam, K. Rajendran, S.K. Lakkaboyana, K. Soontarapa, R. R. R, V.K. Sharma, V. Kumar, K. Venkateswarlu, J.R. Koduru, Recent developments in magnetic nanoparticles and nano-composites for wastewater treatment, Journal of Environmental Chemical Engineering 9(6) (2021) 106553.
[127] E. Kellens, H. Bové, T. Vandenryt, J. Lambrichts, J. Dekens, S. Drijkoningen, J. D'Haen, W.D. Ceuninck, R. Thoelen, T. Junkers, K. Haenen, A. Ethirajan, Micro-patterned molecularly imprinted polymer structures on functionalized diamond-coated substrates for testosterone detection, Biosensors and Bioelectronics 118 (2018) 58-65.
[128] E. Santoso, R. Ediati, Y. Kusumawati, H. Bahruji, D.O. Sulistiono, D. Prasetyoko, Review on recent advances of carbon based adsorbent for methylene blue removal from waste water, Materials Today Chemistry 16 (2020) 100233.
[129] A. Gul, N.G. Khaligh, N.M. Julkapli, Surface modification of Carbon-Based Nanoadsorbents for the Advanced Wastewater Treatment, Journal of Molecular Structure 1235 (2021) 130148.
[130] S. Pacheco, J. Tapia, M. Medina, R. Rodriguez, Cadmium ions adsorption in simulated wastewater using structured alumina–silica nanoparticles, Journal of Non-Crystalline Solids 352(52) (2006) 5475-5481.
[131] H.P. Jarvie, H. Al-Obaidi, S.M. King, M.J. Bowes, M.J. Lawrence, A.F. Drake, M.A. Green, P.J. Dobson, Fate of Silica Nanoparticles in Simulated Primary Wastewater Treatment, Environmental Science & Technology 43(22) (2009) 8622-8628.
[132] W. Ye, L. Liu, B. Zhang, Designing and implementing pollutant emissions trading systems in China: A twelve-year reflection, Journal of Environmental Management 261 (2020) 110207.
[133] C.S. Bezerra, C.M.G. de Farias Lemos, M. de Sousa, L.R.B. Gonçalves, Enzyme immobilization onto renewable polymeric matrixes: Past, present, and future trends, Journal of Applied Polymer Science 132(26) (2015).
[134] M. Afkhami-Ardekani, M.R. Naimi-Jamal, S. Doaee, S. Rostamnia, Solvent-free mechanochemical preparation of metal-organic framework ZIF-67 impregnated by Pt nanoparticles for water purification, Catalysts 13(1) (2022) 9.
[135] M. Martins, C. Mourato, S. Sanches, J.P. Noronha, M.B. Crespo, I.A. Pereira, Biogenic platinum and palladium nanoparticles as new catalysts for the removal of pharmaceutical compounds, Water research 108 (2017) 160-168.
[136] C. Madhusha, T. Jayasundara, I. Munaweera, C. Perera, G. Wijesinghe, M. Weerasekera, C. Sandaruwan, A. Meiyazhagan, F.R. Hernandez, P. Ajayan, Synthesis and structural characterization of copper nanoparticles doped activated carbon derived from coconut coir for drinking water purification, Materials Today Chemistry 27 (2023) 101312.
[137] O.J. Al-sareji, M. Meiczinger, R.A. Al-Juboori, R.A. Grmasha, M. Andredaki, V. Somogyi, I.A. Idowu, C. Stenger-Kovács, M. Jakab, E. Lengyel, K.S. Hashim, Efficient removal of pharmaceutical contaminants from water and wastewater using immobilized laccase on activated carbon derived from pomegranate peels, Scientific Reports 13(1) (2023) 11933.
[138] H. Zeng, L. Zhai, T. Qiao, Y. Yu, J. Zhang, D. Li, Efficient removal of As (V) from aqueous media by magnetic nanoparticles prepared with Iron-containing water treatment residuals, Scientific Reports 10(1) (2020) 9335.

[139] C.D. Powell, A.J. Atkinson, Y. Ma, M. Marcos-Hernandez, D. Villagran, P. Westerhoff, M.S. Wong, Magnetic nanoparticle recovery device (MagNERD) enables application of iron oxide nanoparticles for water treatment, Journal of Nanoparticle Research 22 (2020) 1-11.

[140] Y. Xu, C. Li, X. Zhu, W.E. Huang, D. Zhang, Application of magnetic nanoparticles in drinking water purification, Environmental Engineering & Management Journal (EEMJ) 13(8) (2014).
[141] S.K. Hubadillah, M.H.D. Othman, A. Ismail, M.A. Rahman, J. Jaafar, Y. Iwamoto, S. Honda, M.I.H.M. Dzahir, M.Z.M. Yusop, Fabrication of low cost, green silica based ceramic hollow fibre membrane prepared from waste rice husk for water filtration application, Ceramics International 44(9) (2018) 10498-10509.

[142] E.-R. Kenawya, A.A. Shabakab, A.M. Abou-Zeidb, M.S. Hassounac, M.A. Elhitib, Silica-based nano-adsorbent for enhancement of bacterial biodegradation of methylene blue dye, Desalination and Water treatment 241 (2021) 183-191.
[143] W. Liang, Y. Lu, N. Li, H. Li, F. Zhu, Microwave-assisted synthesis of magnetic surface molecular imprinted polymer for adsorption and solid phase extraction of 4-nitrophenol in wastewater, Microchemical Journal 159 (2020) 105316.
[144] X. Fang, J. Li, B. Ren, Y. Huang, D. Wang, Z. Liao, Q. Li, L. Wang, D.D. Dionysiou, Polymeric ultrafiltration membrane with in situ formed nano-silver within the inner pores for simultaneous separation and catalysis, Journal of Membrane Science 579 (2019) 190-198.
[145] W. Zhang, Z. Zhang, L. Ji, Z. Lu, R. Liu, B. Nian, Y. Hu, Laccase immobilized on nanocomposites for wastewater pollutants degradation: current status and future prospects, Bioprocess and Biosystems Engineering 46(11) (2023) 1513-1531.
[146] S. Datta, R. Veena, M.S. Samuel, E. Selvarajan, Immobilization of laccases and applications for the detection and remediation of pollutants: a review, Environmental Chemistry Letters 19(1) (2021) 521-538.
[147] A.L.P. Guardado, S. Druon-Bocquet, M.-P. Belleville, J. Sanchez-Marcano, A novel process for the covalent immobilization of laccases on silica gel and its application for the elimination of pharmaceutical micropollutants, Environmental Science and Pollution Research 28(20) (2021) 25579-25593.
[148] J. Hou, G. Dong, Y. Ye, V. Chen, Laccase immobilization on titania nanoparticles and titania-functionalized membranes, Journal of Membrane Science 452 (2014) 229-240.
[149] E. Skoronski, D.H. Souza, C. Ely, F. Broilo, M. Fernandes, A.F. Júnior, M.G. Ghislandi, Immobilization of laccase from Aspergillus oryzae on graphene nanosheets, International journal of biological macromolecules 99 (2017) 121-127.
[150] W. Zhou, W. Zhang, Y. Cai, Enzyme-enhanced adsorption of laccase immobilized graphene oxide for micro-pollutant removal, Separation and Purification Technology 294 (2022) 121178.
[151] Y. Tang, W. Li, Y. Muhammad, S. Jiang, M. Huang, H. Zhang, Z. Zhao, Z. Zhao, Fabrication of hollow covalent-organic framework microspheres via emulsion-interfacial strategy to enhance laccase immobilization for tetracycline degradation, Chemical Engineering Journal 421 (2021) 129743.
[152] N. Miletić, A. Nastasović, K. Loos, Immobilization of biocatalysts for enzymatic polymerizations: possibilities, advantages, applications, Bioresource Technology 115 (2012) 126-135.
[153] M. Bilal, T. Rasheed, Y. Zhao, H.M. Iqbal, J. Cui, "Smart" chemistry and its application in peroxidase immobilization using different support materials, International journal of biological macromolecules 119 (2018) 278-290.
[154] T. de Andrade Silva, W.J. Keijok, M.C.C. Guimarães, S.T.A. Cassini, J.P. de Oliveira, Impact of immobilization strategies on the activity and recyclability of lipases in nanomagnetic supports, Scientific Reports 12(1) (2022) 6815.
[155] S. Zahirinejad, R. Hemmati, A. Homaei, A. Dinari, S. Hosseinkhani, S. Mohammadi, F. Vianello, Nano-organic supports for enzyme immobilization: scopes and perspectives, Colloids and Surfaces B: Biointerfaces 204 (2021) 111774.
[156] M. Catauro, S.V. Ciprioti, Characterization of hybrid materials prepared by sol-gel method for biomedical implementations. A critical review, Materials 14(7) (2021) 1788.
[157] L. Lloret, G. Eibes, G. Feijoo, M.T. Moreira, J.M. Lema, F. Hollmann, Immobilization of laccase by encapsulation in a sol–gel matrix and its characterization and use for the removal of estrogens, Biotechnology Progress 27(6) (2011) 1570-1579.
[158] M. Naghdi, M. Taheran, S.K. Brar, A. Kermanshahi-pour, M. Verma, R.Y. Surampalli, Fabrication of nanobiocatalyst using encapsulated laccase onto chitosan-nanobiochar composite, International Journal of Biological Macromolecules 124 (2019) 530-536.
[159] L.S. Queiroz, M. Regnard, F. Jessen, M.A. Mohammadifar, J.J. Sloth, H.O. Petersen, F. Ajalloueian, C.M.C. Brouzes, W. Fraihi, H. Fallquist, A.F. de Carvalho, F. Casanova, Physico-chemical and colloidal properties of protein extracted from black soldier fly (Hermetia illucens) larvae, International Journal of Biological Macromolecules 186 (2021) 714-723.
[160] S.S. Nadar, V.K. Rathod, Magnetic macromolecular cross linked enzyme aggregates (CLEAs) of glucoamylase, Enzyme and Microbial Technology 83 (2016) 78-87.
[161] R. Liu, W. Zhang, S. Wang, H. Xu, Y. Hu, Magnetic Polyethyleneimine Nanoparticles Fabricated via Ionic Liquid as Bridging Agents for Laccase Immobilization and Its Application in Phenolic Pollutants Removal, Molecules 27(23) (2022) 8522.
[162] S. Sadeghzadeh, Z. Ghobadi Nejad, S. Ghasemi, M. Khafaji, S.M. Borghei, Removal of bisphenol A in aqueous solution using magnetic cross-linked laccase aggregates from Trametes hirsuta, Bioresource Technology 306 (2020) 123169.
[163] R. Sarma, M.S. Islam, A.-F. Miller, D. Bhattacharyya, Layer-by-Layer-Assembled Laccase Enzyme on Stimuli-Responsive Membranes for Chloro-Organics Degradation, ACS Applied Materials & Interfaces 9(17) (2017) 14858-14867.
[164] X. Li, Y. Xu, K. Goh, T.H. Chong, R. Wang, Layer-by-layer assembly based low pressure biocatalytic nanofiltration membranes for micropollutants removal, Journal of Membrane Science 615 (2020) 118514.
[165] H.H. Nguyen, M. Kim, An overview of techniques in enzyme immobilization, Applied Science and Convergence Technology 26(6) (2017) 157-163.
[166] J. Chapman, A.E. Ismail, C.Z. Dinu, Industrial applications of enzymes: Recent advances, techniques, and outlooks, Catalysts 8(6) (2018) 238.
[167] W. Huang, W. Zhang, Y. Gan, J. Yang, S. Zhang, Laccase immobilization with metal-organic frameworks: Current status, remaining challenges and future perspectives, Critical Reviews in Environmental Science and Technology 52(8) (2022) 1282-1324.
[168] J.K.H. Wong, H.K. Tan, S.Y. Lau, P.-S. Yap, M.K. Danquah, Potential and challenges of enzyme incorporated nanotechnology in dye wastewater treatment: A review, Journal of environmental chemical engineering 7(4) (2019) 103261.
[169] E. Skoronski, D.H. Souza, C. Ely, F. Broilo, M. Fernandes, A. Fúrigo, M.G. Ghislandi, Immobilization of laccase from Aspergillus oryzae on graphene nanosheets, International Journal of Biological Macromolecules 99 (2017) 121-127.
[170] N.S. Alsaiari, A. Amari, K.M. Katubi, F.M. Alzahrani, H.N. Harharah, F.B. Rebah, M.A. Tahoon, The Biocatalytic Degradation of Organic Dyes Using Laccase Immobilized Magnetic Nanoparticles, Applied Sciences 11(17) (2021) 8216.
[171] X. Chen, B. He, M. Feng, D. Zhao, J. Sun, Immobilized laccase on magnetic nanoparticles for enhanced lignin model compounds degradation, Chinese Journal of Chemical Engineering 28(8) (2020) 2152-2159.
[172] P. Srinivasan, T. Selvankumar, B.A. Paray, M.U. Rehman, S. Kamala-Kannan, M. Govarthanan, W. Kim, K. Selvam, Chlorpyrifos degradation efficiency of Bacillus sp. laccase immobilized on iron magnetic nanoparticles, 3 Biotech 10(8) (2020) 366.
[173] O. Demkiv, G. Gayda, N. Stasyuk, O. Brahinetz, M. Gonchar, M. Nisnevitch, Nanomaterials as Redox Mediators in Laccase-Based Amperometric Biosensors for Catechol Assay, Biosensors 12(9) (2022) 741.
[174] M. Bilal, N. Hussain, J.H.P. Américo-Pinheiro, Y.Q. Almulaiky, H.M.N. Iqbal, Multi-enzyme co-immobilized nano-assemblies: Bringing enzymes together for expanding bio-catalysis scope to meet biotechnological challenges, International Journal of Biological Macromolecules 186 (2021) 735-749.
[175] P. Galliker, G. Hommes, D. Schlosser, P.F.X. Corvini, P. Shahgaldian, Laccase-modified silica nanoparticles efficiently catalyze the transformation of phenolic compounds, Journal of Colloid and Interface Science 349(1) (2010) 98-105.
[176] N.A. Ahmad Jafri, R. A. Rahman, A.H. Mohd Yusof, N.J. Sulaiman, D. Sukmawati, M.S. Mohd Syukri, Adsorption kinetics of immobilized laccase on magnetically-separable hierarchically-ordered mesocellular mesoporous silica, Bioresource Technology Reports 22 (2023) 101445.
[177] J. Zhang, X. Huang, L. Zhang, Y. Si, S. Guo, H. Su, J. Liu, Layer-by-layer assembly for immobilizing enzymes in enzymatic biofuel cells, Sustainable Energy & Fuels 4(1) (2020) 68-79.
[178] V. Semerdzhieva, R. Raykova, D. Marinkova, S. Yaneva, G. Chernev, I. Iliev, Layer-By-Layer Assembly of Enzymes and Nanoparticles onto Cellulose Support, J Biosens Bioelectron 9(263) (2018) 2.
[179] S. Ren, F. Wang, H. Gao, X. Han, T. Zhang, Y. Yuan, Z. Zhou, Recent Progress and Future Prospects of Laccase Immobilization on MOF Supports for Industrial Applications, Applied Biochemistry and Biotechnology (2023).
[180] M.N. Rashed, Adsorption technique for the removal of organic pollutants from water and wastewater, Organic pollutants-monitoring, risk and treatment 7 (2013) 167-194.
[181] A. Bafana, S.S. Devi, T. Chakrabarti, Azo dyes: past, present and the future, Environmental Reviews 19(NA) (2011) 350-371.
[182] C. Zhang, H. Chen, G. Xue, Y. Liu, S. Chen, C. Jia, A critical review of the aniline transformation fate in azo dye wastewater treatment, Journal of Cleaner Production 321 (2021) 128971.
[183] S. Ba, A. Arsenault, T. Hassani, J.P. Jones, H. Cabana, Laccase immobilization and insolubilization: from fundamentals to applications for the elimination of emerging contaminants in wastewater treatment, Critical reviews in biotechnology 33(4) (2013) 404-418.
[184] M. Mazur, P. Krysiński, A. Michota-Kamińska, J. Bukowska, J. Rogalski, G.J. Blanchard, Immobilization of laccase on gold, silver and indium tin oxide by zirconium–phosphonate–carboxylate (ZPC) coordination chemistry, Bioelectrochemistry 71(1) (2007) 15-22.
[185] S.K.S. Patel, H. Choi, J.-K. Lee, Multimetal-Based Inorganic–Protein Hy-

brid System for Enzyme Immobilization, ACS Sustainable Chemistry & Engineering 7(16) (2019) 13633-13638.
[186] Y. Zhu, F. Qiu, J. Rong, T. Zhang, K. Mao, D. Yang, Covalent laccase immobilization on the surface of poly(vinylidene fluoride) polymer membrane for enhanced biocatalytic removal of dyes pollutants from aqueous environment, Colloids and Surfaces B: Biointerfaces 191 (2020) 111025.
[187] N. Ormategui, A. Veloso, G.P. Leal, S. Rodriguez-Couto, R. Tomovska, Design of Stable and Powerful Nanobiocatalysts, Based on Enzyme Laccase Immobilized on Self-Assembled 3D Graphene/Polymer Composite Hydrogels, ACS Applied Materials & Interfaces 7(25) (2015) 14104-14112.
[188] Y.-S. Zimmermann, P. Shahgaldian, P.F.X. Corvini, G. Hommes, Sorption-assisted surface conjugation: a way to stabilize laccase enzyme, Applied Microbiology and Biotechnology 92(1) (2011) 169-178.
[189] R.A. Fernandes, A.L. Daniel-da-Silva, A.P.M. Tavares, A.M.R.B. Xavier, EDTA-Cu (II) chelating magnetic nanoparticles as a support for laccase immobilization, Chemical Engineering Science 158 (2017) 599-605.
[190] M. Fernández-Fernández, M.Á. Sanromán, D. Moldes, Recent developments and applications of immobilized laccase, Biotechnology Advances 31(8) (2013) 1808-1825.