

## **A REVIEW: FACILE AND GREEN SYNTHESIS OF MARINE MACROALGAE AND ITS PHOTOCATALYTIC PERFORMANCE ON POLLUTED WATER REMEDIATION**

### **REVIEW: SINTESIS HIJAU DAN MUDAH MAKROALGA LAUT DAN KINERJA FOTOKATALITIKNYA PADA REMEDIASI AIR BERPOLUTAN**

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#### **Abstrak**

Ulasan artikel ini mengulas kemajuan terbaru dalam sintesis hijau dan mudah nano-partikel logam dan oksida logam menggunakan beragam makroalga laut. Banyak penelitian telah menunjukkan biosintesis perak, emas, oksida besi, seng oksida, dan nanopartikel lainnya melalui teknik "*phycosynthesis*" berkelanjutan tanpa bahan kimia beracun dengan memanfaatkan biomolekul makroalga sebagai reduktan dan stabilisator. Nanopartikel yang dimediasi alga ini menunjukkan aktivitas fotokatalitik yang penting dalam mendegradasi berbagai pewarna organik dan polutan yang diindikasikan dari morfologi dan sifatnya. Telaah artikel menunjukkan nanopartikel yang dihasilkan dari makroalga seperti *Sargassum horneri*, *Syringodium isoetifolium* dan *Ulva lactuca* mampu mencapai efisiensi degradasi lebih dari 90% terhadap kontaminasi pewarna seperti metilen biru, metil oranye, dan rhodamin B. Mekanisme ini dihipotesiskan melibatkan foto-eksitasi yang menghasilkan spesies oksidatif reaktif pada permukaan nanopartikel yang dengan cepat membelah molekul pewarna. Peningkatan pemahaman tentang peran biomolekul makroalga dan optimalisasi kondisi sintesis dapat memungkinkan peningkatan kinerja katalitik. Dengan demikian, hasil dari literatur yang dikumpulkan dan disajikan dalam studi ini menunjukkan potensi makroalga laut yang sangat menjanjikan untuk degradasi fotokatalitik dan remediasi air lingkungan. Dengan ketersediaan luas dan keanekaragaman hayati yang sebagian besar belum dimanfaatkan, makroalga laut terbukti menjanjikan sebagai sumber daya terbarukan untuk mengembangkan teknologi remediasi air ramah lingkungan melalui desain nanokatalis logam / oksida logam.

*Kata Kunci: Sintesis hijau, Makroalga laut, Nanopartikel logam, Fotokatalisis, Remediasi air tercemar*

#### **Abstract**

This review critically examines the burgeoning field of research focused on the environmentally benign synthesis of metallic nanoparticles exploiting the unique properties and diverse array of marine macroalgae species. Some studies have discovered the biosynthesis of silver, gold, iron oxide, zinc oxide, and other nanoparticles through a sustainable "*phycosynthesis*" technique without toxic chemicals with utilize the macroalgal biomolecules as reductants and stabilizers. These algae-mediated nanoparticles exhibit notable photocatalytic activities in degrading various organic dyes and pollutants that indicated with their morphologies and properties. The article review showed that nanoparticles produced from macroalgae such as *Sargassum horneri*, *Syringodium isoetifolium* and *Ulva lactuca* can reach over than 90% degradation efficiency to contamination of dyes like methylene blue, methyl orange, and rhodamine B. The mechanism is hypothesized to involve photoexcitation generating reactive oxidative species on the nanoparticle surface that rapidly cleave dye molecules. Improved understanding of the roles of macroalgal biomolecules and optimization of synthesis conditions may enable enhanced catalytic performances. Herein, the result from the literatures collected and presented in these studies demonstrates highly promising potential of marine macroalgal for photocatalytic degradation and environmental water remediation. With their widespread availability and biodiversity remaining largely untapped, marine macroalgae prove promising as renewable resources for developing eco-friendly water remediation technologies through metal/metal oxide nanocatalyst design.

*Keywords: Green synthesis, Marine macroalgae, Metal nanoparticles, Photocatalysis, Polluted water remediation*

## INTRODUCTION

Polluted water instigated by industrial effluents and domestic waste comprehending heavy metals, dye, and organic pollutants is an escalating environmental concern. Conventional water treatment methods are often ineffective and generate secondary waste. Photocatalysis employing sustainable materials has emerged as a promising solution for polluted water remediation (Chauke et al., 2024).

The oceans contain vast potential wealth, not only in aesthetic beauty but also in natural resources that can provide tangible solutions to pressing human needs. One key area is environmental remediation of pollutants, which threatens ecosystems and human health. A promising technology that offers efficient and cost-effective solutions is nanoparticles. As an expansive, interdisciplinary field, nanoparticles derived from marine sources have generated growing research interest.

The oceans are home to macroalgae that serve as renewable and sustainable green resources (Sudhakar et al., 2018). Because of this reason, a prominent interest in using such algae as sources for synthesizing nanoparticles for environmental applications is increasing. With rich biodiversity that remains largely untapped, the world's oceans may offer new alternative to synthesized and produces green nanoparticles and other solutions to counteract environmental pollution.

Recently, marine macroalgae have attracted significant research attention as promising green resources for producing metal/metal oxide nanoparticles through a facile 'phycosynthesis' process, without using toxic chemicals. Marine macroalgae are a diverse group of multicellular algae found abundantly in oceans (Leandro et al., 2020). Their intrinsic biomolecules allow for controlled nanoparticle fabrication via phyco-synthesis (Chaudhary et al., 2020).

Numerous investigations have been conducted to elucidate the potential of diverse marine macroalgal genera as viable biogenic systems for the controlled synthesis of various metallic nanoparticles, encompassing gold (AuNPs), silver (AgNPs), iron (FeNPs), zinc oxide (ZnONPs), and others. These biogenically derived nanoparticles have exhibited promising photocatalytic performance, underscoring the efficacy of these sustainable synthesis routes leveraging marine biomaterials. A key attribute is marine macroalgae providing natural stabilizing and capping agents during phyco-synthesis (Kim et al., 2021), yielding stable nanocatalysts/algal nanoparticles (NPs) for remediation in wastewater containing dye.

However, further optimization is yet indispensable to effusively comprehend and

utilize the photocatalytic potential of macroalgae-mediated nanomaterials. This review aims to survey recent advances in facile phyco-synthesis approaches utilizing various marine macroalgae. It will discuss the roles of marine macroalgal nanoparticles in several environmental application and photocatalytic performance for polluted water remediation. Insights could guide improved understanding bio-photocatalyst design and selection of algal species to address this global challenge.

## NANOPARTICLES (NPs): AN OVERVIEW

Nanotechnology, a transformative field at the nexus of materials science and engineering, hinges on the precise manipulation of matter at the nanoscale, typically defined as dimensions ranging from 1 to 100 nanometers (Rizvi et al., 2022). This meticulous control over matter at such minuscule scales unlocks the creation of nanoparticles (Joudeh & Linke, 2022), endowed with unique physicochemical properties that diverge significantly from their bulk counterparts (Patil & Chandrasekaran, 2020). These emergent properties arise from the inherently high surface area-to-volume ratio and quantum confinement effects characteristic of the nanoscale, rendering nanoparticles exquisitely sensitive to surface interactions and enabling enhanced reactivity and tunable optical and electronic properties (Altammar, 2023; Joudeh & Linke, 2022; Rizvi et al., 2022). This remarkable versatility has positioned nanoparticles at the forefront of innovation across a multitude of disciplines, including medicine, where they hold promise as targeted drug delivery vehicles (Almurshedi et al., 2023) and imaging agents (Jameel et al., 2021); electronics, where their unique optical and electronic properties are harnessed in high-performance sensors (Ibrahim et al., 2024; Srikhao et al., 2022) and displays (Dhar et al., 2020); and environmental science, where they offer effective solutions for remediation and pollution control (Varghese Alex et al., 2020). Broadly categorized as either inorganic or organic, nanoparticles encompass a diverse library of materials, each with tailored properties and applications dictated by their composition, size, shape, and surface chemistry (Khan et al., 2022).

Inorganic nanoparticles, encompassing a diverse class of materials including metals and metal oxides, have garnered significant attention due to their unique physicochemical properties and their resulting applications (Ciobotaru et al., 2023). Metal nanoparticles, with gold (Kaithavelikkakath Francis et al., 2020), silver (Algotiml et al., 2022), and copper (Shaikhaldein et al., 2024) being prominent examples, are often synthesized through methods such as chemical

reduction (Ansari et al., 2023; Singh et al., 2018)), where metal ions in solution are reduced to their zero-valent state, or physical vapor deposition, where metals are vaporized and subsequently condensed onto a substrate. These nanoparticles exhibit distinctive optical properties, often arising from localized surface plasmon resonance, a phenomenon characterized by the collective oscillation of electrons in response to incident light (Petryayeva & Krull, 2011).

Gold nanoparticles, for example, are highly sought after for biomedical applications, including drug delivery systems (Sulaiman et al., 2020) and biosensing (Ghoniem et al., 2023), due to their biocompatibility and tunable plasmonic properties, which allow for targeted delivery and sensitive detection. Silver nanoparticles, renowned for their potent antimicrobial activity (X. Liu et al., 2021), are utilized in wound dressings to promote healing (Sulastri et al., 2023) and in water purification systems (Che et al., 2019) to eliminate harmful bacteria.

Metal oxide nanoparticles, on the other hand, have found widespread applications in diverse fields ranging from catalysis to medicine. Titanium dioxide (TiO<sub>2</sub>), a wide-bandgap semiconductor, is synthesized through methods such as sol-gel processing or hydrothermal synthesis, which allow for control over particle size and morphology (Gatou et al., 2024). Its strong UV absorption properties have led to its widespread incorporation into sunscreens, where it acts as a physical blocker of harmful ultraviolet radiation (Aguilera et al., 2023; Smijs & Pavel, 2011). Zinc oxide (ZnO), another metal oxide with notable antimicrobial properties (Mendes et al., 2022), is incorporated into cosmetics (Mohammed et al., 2023) and textiles (Shaban et al., 2018) to impart antibacterial and odor-control properties (Tania et al., 2022).

Organic nanoparticles, synthesized from organic molecules, offer a compelling platform for biomedical applications due to their inherent biocompatibility, often exhibiting low toxicity profiles, and their capacity for biodegradation, mitigating concerns of long-term accumulation in biological systems (Khan et al., 2022).

Polymer nanoparticles, crafted from a wide range of polymers, including synthetic polymers such as polystyrene (Jung et al., 2023), polyethylene glycol (PEG) (Jokerst et al., 2011), and polylactic acid (PLA) (Ekinci et al., 2022), as well as natural polymers like chitosan (Van Bavel, Issler, et al., 2023) and alginate (Thai et al., 2020; Van Bavel, Lewrenz, et al., 2023), are typically synthesized through techniques such as emulsion polymerization, where monomers are polymerized within dispersed droplets, or nanoprecipitation, where a rapid change in solvent conditions induces polymer aggregation

into nanoparticles (Zielińska et al., 2020). Their versatility, coupled with the ability to fine-tune their size, surface charge, and drug release profiles through modifications in polymer composition and synthesis parameters, has led to their widespread exploration as drug delivery vehicles (Beach et al., 2024), enhancing drug solubility, prolonging circulation time, and enabling targeted delivery to specific cells or tissues (Mitchell et al., 2021).

Dendrimers, a unique class of polymeric nanoparticles, possess a well-defined branched architecture emanating from a central core, providing them with a high density of surface functional groups (Pérez-Ferreiro et al., 2023). Synthesized through a stepwise chemical process, dendrimers offer precise control over size, shape, and surface functionality (Walter & Malkoch, 2012), making them attractive for applications in drug delivery (An et al., 2023) and imaging (Cruz et al., 2023; J. Liu et al., 2019). Their unique structure allows for high drug loading capacities, while their surface functional groups can be tailored for targeted delivery and enhanced cellular uptake (Lyu et al., 2020).

Liposomes, spherical vesicles composed of a phospholipid bilayer surrounding an aqueous core, are assembled through methods such as lipid film hydration or reverse phase evaporation (Akbarzadeh et al., 2013; Lombardo & Kiselev, 2022). Their structural similarity to cell membranes renders them highly biocompatible and well-suited for drug delivery applications (Arul et al., 2023), capable of encapsulating both hydrophilic drugs within their aqueous core (Mehta et al., 2023) and hydrophobic drugs within their lipid bilayer (Tenchov et al., 2021), thereby improving drug solubility, reducing toxicity, and enabling controlled release (Nsairat et al., 2022).

Herein nanotechnology, with its diverse toolkit of nanoparticles exhibiting a remarkable spectrum of properties, stands poised to revolutionize numerous sectors, holding particular promise in addressing the pressing environmental challenges of our time. Metal and metal oxide nanoparticles, in particular, are emerging as powerful allies in the quest for effective environmental remediation strategies. Iron nanoparticles, for instance, with their inherently high surface area and reactivity, are proving to be exceptionally adept at degrading and removing persistent organic pollutants from contaminated water sources. Their efficacy stems from their ability to readily donate electrons, driving redox reactions that break down harmful pollutants such as pesticides (Rani et al., 2023), industrial solvents (Muthukumar et al., 2024; Twinkle et al., 2024), and chlorinated hydrocarbons (Gil-Díaz et al., 2022) into less toxic

byproducts. Similarly, titanium dioxide (TiO<sub>2</sub>) nanoparticles, renowned for their photocatalytic prowess (D. Bristow et al., 2024), are being strategically deployed in wastewater treatment plants to degrade harmful organic pollutants. Upon exposure to sunlight, these nanoparticles act as miniature photocatalytic reactors, generating reactive oxygen species (ROS) that effectively break down a wide range of organic contaminants, including dyes, pharmaceuticals, and endocrine disruptors, into less harmful substances. The ongoing exploration and refinement of these and other nanomaterials, coupled with the development of even more efficient, targeted, and environmentally benign synthesis methods, hold immense promise for creating a cleaner, more sustainable future. As our understanding of nanoparticle design, behavior, and environmental fate advances, we can harness the transformative power of nanotechnology to address pressing environmental challenges and pave the way for a healthier planet.

#### MARINE MACROALGAE FOR GREEN SYNTHESIS OF NANOPARTICLES

Green and sustainable synthesis techniques have been successfully utilized to bio-fabricate various metal nanoparticles from a variety of

marine macroalgae species. These metal nanoparticles synthesized using algal biomass demonstrate notable applications in photocatalytic degradation of organic dyes. It is worth mentioning that extensive research has been carried out in this area owing to the promptly emergent domain of green and novel material for environmental remediation which focuses on employing eco-friendly approaches for nanoparticle production. A plethora of investigations has elucidated the efficacious biosynthesis of silver nanoparticles (AgNPs) employing aqueous extracts derived from diverse species of marine macroalgae, encompassing *Ulva lactuca*, *Sargassum myriocystum*, and *Caulerpa racemosa*.

Table 1 illustrates the many aquatic macroalgae species that have been thoroughly investigated for their potential in photocatalytic degradation processes and the manufacture of metal nanoparticles. The inherent constitution of marine macroalgae, replete with an array of bioactive moieties, including proteins, phenolic compounds, and polysaccharides, which exhibit reducing and capping capabilities, has rendered them a propitious source of biomaterials for the eco-friendly synthesis of metallic nanoparticles (Edison et al., 2016; Ramakrishna et al., 2016).

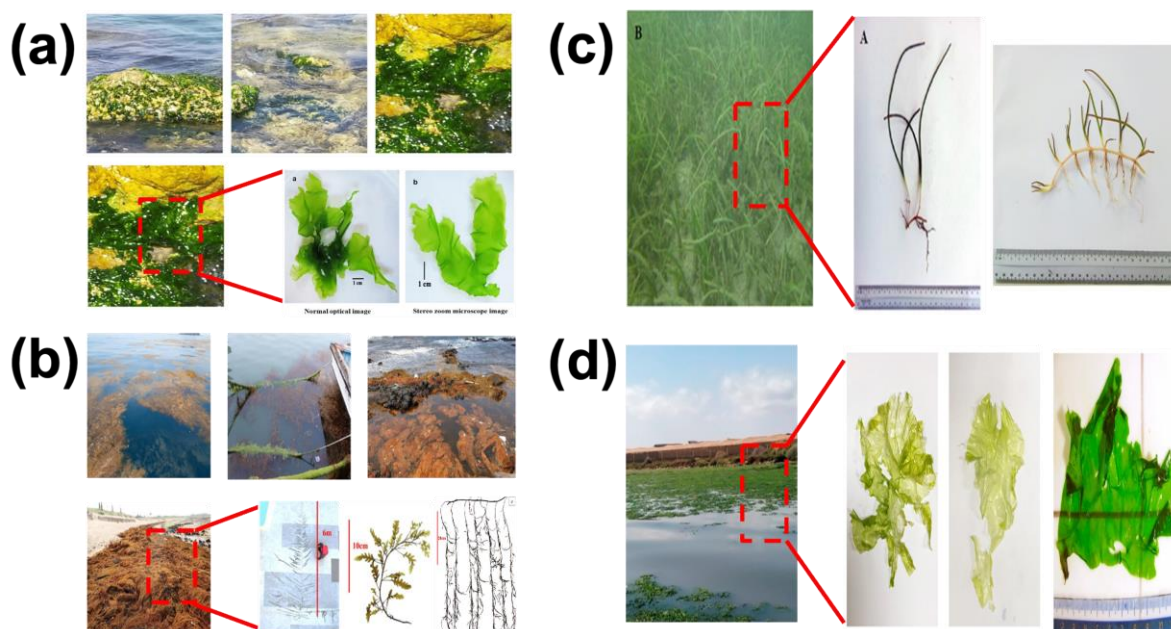


Figure 1. The marine macroalgae, (a) *Ulva fasciata* on Egyptian sea shoreline and assessment of their morphological via direct visual observation and by using the microscope, (b) *Sargassum horneri* on Jeju Province, South Korea shoreline and assessment of their morphological via direct visual observation, (c) *Syringodium isoetifolium* on Myanmar coastal area of Rakhine province, and (d) *Ulva lactuca* on Mediterranean sea coastal lagoon (Marchica lagoon) in Morocco (adopted and modified from (a) El-Gendy et al. (2023), (b) Byeon et al. (2020) and Wu et al. (2023), (c) Thu-Rein et al. (2019) and Soe-Htun et al. (2017), and (d) García-Poza et al. (2022); Hamouda et al. (2023) and Rahhou et al. (2023) is licensed under [CC BY 4.0](https://creativecommons.org/licenses/by/4.0/)).

Table 1. Species of Marine Macroalgae Used for Phycosynthesis and Applications of Metal Nanoparticles for Photocatalytic Degradation

Species of Algae	NPs synthesized and produced	Size of NPs (nm)	Shape of NPs	Applications	Reference
<i>Caulerpa racemosa</i>	Ag	Approx. to 25	Distorted spherical	Catalytic degradation of MB	(Edison et al., 2016)
<i>Turbinaria conoides</i> & <i>Sargassum tenerrimum</i>	Au	27-35	Nearly spherical	Catalysts for the reduction of aromatic nitro compounds and organic dye molecules	(Ramakrishna et al., 2016)
<i>Hypnea musciformis</i>	Ag	2-55.8	Well-separated spherical	Photocatalytic degradation of methyl orange dye	(Ganapathy Selvam & Sivakumar, 2015)
<i>Sargassum vulgare</i>	Zn	50-150	Spherical	Degradation of methylene blue dye under UV light irradiation	(Karkhane et al., 2020)
<i>Sargassum myriocystum</i>	Ag	20 ± 2.2	Well dispersed hexagonal	Photocatalytic activity of methylene blue (MB) degradation	(Balaraman et al., 2020)
<i>Ulva lactuca</i>	Ag	48.59	Spherical	Photocatalytic degradation of methyl orange dye	(Kumar et al., 2013)
<i>Sargassum coreanum</i>	Ag	19	Distorted spherical shape	Catalytic degradation of methylene blue	(Somasundaram et al., 2021)
<i>Padina gymnospora</i>	CdO-ZnO (SCZ)	20-50	Distorted hexagonal	Photocatalytic degradation activity of Reactive Blue 198 dye under the UV light, visible light and natural sunlight irradiation	(Rajaboopathi & Thambidurai, 2017)
<i>Ulva lactuca</i>	Ag	Approx. to 24	Spherical	Antibacterial activity	(Gurusamy et al., 2019)

The investigation conducted by Edison et al. (2016) elucidated the facilitated extracellular biosynthesis of silver nanoparticles (AgNPs) exhibiting an average diameter of 25 nm and a deformed spherical morphology, employing *Caulerpa racemosa*. These AgNPs manifested exceptional catalytic activity in the photodegradation of methylene blue (MB). Concurrently, Ramakrishna et al. (2016) reported the collaborative synthesis of nearly spherical gold nanoparticles (AuNPs) with dimensions ranging from 27 to 35 nm, utilizing the synergistic action of *Turbinaria conoides* and *Sargassum tenerrimum*. The resultant AuNPs demonstrated efficacious photocatalytic properties in the reduction of organic dyes and aromatic nitro compounds.

*Hypnea musciformis* mediated AgNPs formation ranging from 2-55.8 nm with well-separated spherical configuration displaying photocatalytic degradation of MO dye under visible light (Ganapathy Selvam & Sivakumar, 2015). *Sargassum vulgare* led to the biosynthesis of ZnONPs measuring 50-150 nm with spherical

shape, which catalyzed the photodegradation of MB dye under ultraviolet irradiation (Karkhane et al., 2020). Similarly, *Sargassum myriocystum* assisted production of hexagonal AgNPs averaging 20 ± 2.2 nm exhibited photocatalytic activity for methylene blue degradation (Balaraman et al., 2020).

*Ulva lactuca* facilitated green biosynthesis of spherical shaped AgNPs measuring 48.59 nm with distinct photocatalytic activity towards methyl orange dye degradation (Kumar et al., 2013). Likewise, AgNPs formed by *Sargassum coreanum* measured around 19 nm with distorted spherical morphology catalyzing the photodegradation of methylene blue dye (Somasundaram et al., 2021). The investigation conducted by Rajaboopathi and Thambidurai (2017) demonstrated the mediation of *Padina gymnospora* in the synthesis of composite cadmium oxide-zinc oxide nanoparticles (SCZNPs). These nanoparticles exhibited a size distribution ranging from 20 to 50 nm and a distorted hexagonal morphology. Notably, the SCZNPs manifested photocatalytic degradation

capabilities toward Reactive Blue 198 dye under diverse illumination sources. Additionally, *Ulva lactuca* assisted production of AgNPs around 24 nm exhibited strong antibacterial efficacy (Gurusamy et al., 2019).

Marine macroalgae possess tremendous potential as eco-friendly nanofactories for producing diverse NPs with distinct morphological attributes and applications in photocatalysis and antibacterial activities as systematically demonstrated in this review. Comprehensively, the detailed analysis covering species-specific NP biosynthesis and applications provides valuable insights towards the development of sustainable nanotechnologies.

### PHYCOSYNTHESIS METHOD

The study by Kim et al. (2021) conveyed the preparation of macroalgal extract of *Sargassum serratifolium* (SS) and its green synthesis of *Sargassum serratifolium* (SS-AgNPs) as illustrated by Figure 2. In the same manner, the exploration by Song et al. (2022) successfully utilized *Sargassum horneri* extract in application of toxic azo dye catalytic degradation. This process starts from collected the macroalgae *Sargassum serratifolium* (SS) from the sea sites. After that, the fresh macroalgae was thoroughly blended into a subtle powder using a blender (Song et al., 2022). To prepare the algal extract, the fine pulverize form of SS was dissolved in ethanol with concentration of 80% at room temperature (Kim et al., 2021; Song et al., 2022). Then, the mixture was purified to remove insoluble particles. The filtrate then concentrated by removing the solvent using a rotary vacuum evaporator. This produced a viscous extract of SS which was freeze-dried into powder form. For using the dried SS powder to biofabricate NPs in the future, it was stored at 75°C. To initiate the nanoparticle synthesis procedure, the powder was solubilized in an aqueous medium at a concentration of 4 mg/mL. Prior to commencing the phycosynthesis process, this solution underwent filtration through a 0.2 µm syringe filter to eliminate any potential spore contamination (Song et al., 2022).

The illustration depicted in Figure 2(b) elucidates an environmentally benign approach for the synthesis of silver nanoparticles (AgNPs) utilizing *Sargassum* species (SS) extracts as reducing and stabilizing agents. On the left side, the precursor solutions are portrayed, comprising silver nitrate (AgNO<sub>3</sub>) and the algal extract of SS, which contains phytochemical compounds that function as stabilizing agents. These solutions are amalgamated in specific ratios and subsequently subjected to thermal

treatment at 80°C for a duration of 15 minutes, as indicated by the temperature and time settings on the heating device situated in the center. The investigation by Sreebamol et al. (2023) elucidate that during this heating process, the biomolecules present in the plant extract serve as reducing agents, facilitating the conversion of silver ions (Ag<sup>+</sup>) to metallic silver (Ag<sup>0</sup>) nanoparticles. Concomitantly, these biomolecules also stabilize and cap the synthesized AgNPs, thereby preventing agglomeration (Rezazadeh et al., 2020; Sundaresan & Kasi, 2023; Vinayagam et al., 2024). The resultant AgNPs solution can then be characterized employing techniques such as UV-Vis spectroscopy, scanning electron microscopy (SEM), and transmission electron microscopy (TEM) (Solanki & Patel, 2023). By using the same principle of green fabrication, it is numerous reported in literature of efficacious utilization of macroalgal extract to green nanoparticles (Balaraman et al., 2020; Fouda et al., 2022; Ganapathy Selvam & Sivakumar, 2015; Khalafi et al., 2019; Soleimani et al., 2023).

This environmentally-friendly green synthesis method avoids harsh chemicals and leverages the reducing and stabilizing capabilities of marine macroalgal extracts to produce NPs with potential applications across biomedical (Machado et al., 2021), catalytic (Alarfaj et al., 2024), and optoelectronic fields (Vinayagam et al., 2024).

Prior to the study of Kim et al. (2021), the biosynthesis of silver and gold nanoparticles (AgNPs and AuNPs) necessitated the optimization of several parameters, including temperature, reaction time, concentration of *Sargassum serratifolium* extract, and the metal precursors employed. For the synthesis of AgNPs, 1 mL of the filtered *S. serratifolium* extract was amalgamated with 1 µL of a 1 M aqueous silver nitrate solution, resulting in a final concentration of 2 mg/mL. This mixture was subsequently incubated at 80°C for a duration of fifteen minutes. To terminate the reaction, the tube containing the colloidal solution was promptly immersed in an ice bath for five minutes. The formation of SS silver nanoparticles was confirmed by the visual observation of a color transition from yellow to dark orange. Regarding AuNP synthesis, 1 µL of 1 M gold chloride trihydrate solution was added to 1 mL filtered *S. serratifolium* extract (2 mg/mL) and subjected to the same reaction conditions as for AgNPs. Here, the color transformation to violet verified the successful synthesis process of SS-AuNPs which had been biosynthesized successfully (Song et al., 2022).

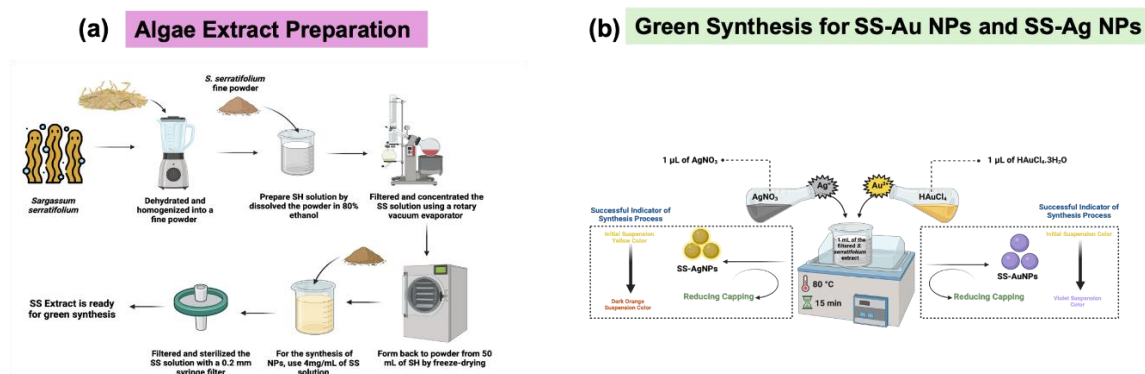


Figure 2. The schematic representation of (a) preparation of SS extract for green synthesis of NPs and (b) The procedure of bio-fabrication of SS-Au NPs and SS-Ag NPs (where the marine macroalgal of SS during the formation of NPs extract acted as stabilizing agents) (adopted and modified from Kim et al. (2021) is licensed under [CC BY 4.0](https://creativecommons.org/licenses/by/4.0/))

Table 2. Macroalgae-NPs application for dye pollutant removal from water and wastewater

Species of Algae	NPs synthesized and produced	Dye removed	Removal efficiency (%)	Reference
<i>Syringodium isoetifolium</i>	TiO <sub>2</sub>	Methylene blue	83	(Sundar et al., 2024)
<i>Syringodium isoetifolium</i>	TiO <sub>2</sub>	Methylene orange	58	(Sundar et al., 2024)
<i>Sargassum horneri</i>	Ag	Methylene blue	NA**	(Song et al., 2022)
<i>Sargassum horneri</i>	Ag	Rhodamine B	NA**	(Song et al., 2022)
<i>Sargassum horneri</i>	Ag	Methyl Orange	NA**	(Song et al., 2022)
<i>Sargassum horneri</i>	Au	Methylene blue	NA**	(Song et al., 2022)
<i>Sargassum horneri</i>	Au	Rhodamine B	NA**	(Song et al., 2022)
<i>Sargassum horneri</i>	Au	Methyl Orange	NA**	(Song et al., 2022)
<i>Caulerpa racemoedisosa</i>	Ag	Methylene Blue	NA*	(Edison et al., 2016)
<i>Turbinaria conoides</i> and <i>Sargassum tenerrimum</i>	Au	Organic Dye Molecules	NA*	(Ramakrishna et al., 2016)
<i>Hypnea musciformis</i>	Ag	Methyl Orange	NA*	(Ganapathy Selvam & Sivakumar, 2015)
<i>Sargassum vulgare</i>	Zn	Methylene Blue	87.59	(Karkhane et al., 2020)

Species of Algae	NPs synthesized and produced	Dye removed	Removal efficiency (%)	Reference
<i>Sargassum myriocystum</i>	Ag	Methyl Orange	98	(Balaraman et al., 2020)
<i>Ulva lactuca</i>	Ag	Methylene Blue	NA*	(Kumar et al., 2013)
<i>Sargassum coreanum</i>	Ag	Reactive Blue 198	>99	(Somasundaram et al., 2021)
<i>Padina gymnospora</i>	CdO-ZnO (SCZ)	Methyl Orange	99.57, 95.2 and 94.3	(Rajaboopathi & Thambidurai, 2017)
<i>Ulva fasciata</i> Delile	ZnO	Methylene Blue	84.9 ± 1.2 (under UV light) and 53.4 ± 0.7 (under the dark)	(Fouda et al., 2022)
<i>Ulva fasciata</i> Delile	ZnO	Tanning Wastewater	96.1 ± 1.7	(Fouda et al., 2022)

Note: NA\*= Not Available, NA\*\*= The authors concludes that the removal efficiency as extremely effective

### MARINE MACROALGAE-NPs FOR DYE POLLUTED WATER REMEDIATION APPLICATION

As shown by Table 2, numerous studies have reported the successful use of metal nanoparticles biosynthesized via marine macroalgae for competent photodegradation of various toxic organic dyes and wastewaters. Sundar et al. (2024) documented the biosynthesis of TiO<sub>2</sub> NPs using *Syngodium isoetifolium* which exhibited excellent photocatalytic removal of methylene blue (83%) and methylene orange (58%) dyes. Likewise, Song et al. (2022) evaluated the photocatalytic performance of AgNPs and AuNPs produced via *Sargassum horneri* against MB, rhodamine B and MO dyes.

Numerous other studies have documented the efficacious photodegradation of toxic dyes employing metal nanoparticles biosynthesized with the aid of macroalgal biomass. For instance, Edison et al. (2016) and Ramakrishna et al. (2016) reported on the biofabrication of AgNPs and AuNPs mediated by the biomass of *Caulerpa racemosa* and the synergistic action of *Turbinaria conoides* and *Sargassum tenerrimum*, respectively. Concurrently, Ganapathy Selvam & Sivakumar (2015) elucidated the biosynthesis of AgNPs facilitated by *Hypnea musciformis* and their activity against methyl orange dye. Additionally, Karkhane et al. (2020) studied photocatalytic performance of ZnONPs mediated by *Sargassum vulgare* for methylene blue degradation. Balaraman et al. (2020) evaluated AgNPs biosynthesis via *Sargassum myriocystum* against methyl orange achieving 98% removal. Likewise, Kumar et al. (2013), Somasundaram et al. (2021) and Rajaboopathi & Thambidurai

(2017) explored dye degradation potential of AgNPs, AgNPs and CdO-ZnO NPs respectively using *Ulva lactuca*, *Sargassum coreanum* and *Padina gymnospora* as algal biomass.

Furthermore, recent studies reported excellent photocatalytic activities of ZnO NPs toward methylene blue dye and tanning wastewater degradation achieving 84.9-96.1% removal efficiency using *Ulva fasciata* biomass (Fouda et al., 2022). Collectively, these findings highlight the promising application of algal-mediated metal nanoparticles in wastewater treatment through photocatalytic processes.

### PHOTODEGRADATION MECHANISM FOR DYE POLLUTED WATER BY MARINE MACROALGAE-NPs

The study conducted by Somasundaram et al. (2021) employed the marine macroalga *Sargassum coreanum* as a biofactory for the synthesis of gold nanoparticles (SC-AgNPs), which exhibited remarkably effective photocatalytic dye degradation capabilities. Figure 3 presents a general possible mechanism for the photodegradation of methylene blue (MB) in the presence of the SC-AgNPs. In aqueous solutions, methylene blue is typically stabilized in its colorless, reduced form (leuco-methylene blue, LMB). Conversely, in an oxidized state, methylene blue assumes a blue coloration and can be readily reduced to colorless degradation products by reducing agents. The breakdown of methylene blue by the silver nanoparticles likely follows a Langmuir-Hinshelwood kinetic model. Within this system, sodium borohydride acts as both an electron donor and a hydrogen source. The silver nanoparticles facilitate the transfer of

electrons from the borohydride ions to the methylene blue molecules owing to the highly negative reduction potential of silver. Upon addition of sodium borohydride to a solution containing methylene blue and silver nanoparticles, borohydride ions and methylene blue molecules adsorb onto the silver

nanoparticle surfaces. This allows rapid electron and hydrogen transport between the adsorbed species. Desorption of the colorless degraded products then occurs, which may free up additional catalytic sites on the nanoparticles for further methylene blue breakdown given the extensive surface area of the nanoparticles.

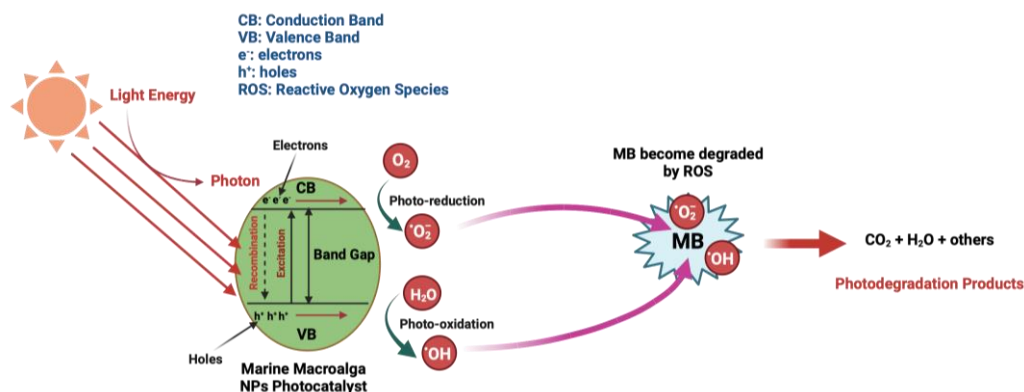


Figure 3. Plausible mechanism of Methylene Blue (MB) photodegradation by using the marine macroalgal-NPs as result of green phycosynthesis process

However, in many cases, the plausible mechanism of dye photodegradation by marine macroalgal-NPs can be simply explained from the nanoparticles act as light-activated semiconductors, generating electron-hole pairs upon irradiation as shown by Figure 3. Furthermore, the general degradation mechanism of action by algal-NPs in many instances mirrors a light-activated semiconductor process. Upon irradiation, these nanoparticles, often enriched with bioactive compounds such as polysaccharides (Abdel-Raouf et al., 2019) and phenolic (Ponnusamy et al., 2024) compounds derived from their algal source, undergo photoexcitation, generating electron-hole pairs. This photocatalytic activity drives dye degradation through several key pathways. Firstly, the photogenerated electrons can directly reduce dissolved oxygen, forming highly reactive superoxide radicals, while the holes oxidize water molecules, producing hydroxyl radicals. These potent reactive oxygen species then initiate a cascade of oxidative reactions that effectively cleave the dye molecules, breaking them down into smaller, less harmful fragments (Anjali et al., 2022). Secondly, the algal nanoparticles, with their inherently high surface area, serve as efficient adsorbents for dye molecules (Osorio-Aguilar et al., 2023), concentrating them near the catalyst surface and facilitating further degradation. This adsorption process, governed by interactions like van der Waals forces and hydrogen bonding (Mansour et al., 2022), effectively isolates the dye molecules, promoting their interaction with the

photogenerated reactive species and accelerating their breakdown. Finally, the inherent biomolecules present in the algal extracts, such as polysaccharides (Abdel-Raouf et al., 2019) and proteins (Sreebamol et al., 2023), likely act as stabilizing agents, preventing nanoparticle aggregation and thereby ensuring sustained catalytic activity over extended periods (Maduraimuthu et al., 2023). In essence, this synergistic interplay of mechanisms encompassing direct redox reactions driven by photogenerated electrons and holes, enhanced adsorption due to the high surface area of the nanoparticles, and stabilization by algal-derived compounds underscores the potential of marine macroalgal nanoparticles as sustainable, cost-effective, and highly effective agents for dye degradation and environmental remediation field.

## CONCLUSION

This review has highlighted the promising potential of utilizing various species of marine macroalgae such as *Sargassum horneri*, *Syringodium isoetifolium* and *Ulva lactuca* for the green synthesis of metal/metal oxide nanoparticles via a facile phycosynthesis route. The result from the literatures collected and presented in these studies demonstrates highly promising potential of marine macroalgal for photocatalytic degradation and environmental water remediation. Moreover, extensive studies have also proven the efficacy of nanoparticles produced with macroalgal biomass for catalyzing

the photodegradation of several toxic-organic dyes and pollutants in wastewater. Key attributes such as intrinsic biomolecules acting as reducing and stabilizing agents yield stable nanocatalysts with good photocatalytic performance. While research efforts have focused on synthesizing silver, gold, iron oxide and zinc oxide nanoparticles, further exploration of other metal/metal oxide systems is warranted. Optimization of phycosynthesis conditions may help enhance catalytic activity. Insights gained into selectivity of different macroalgal genera can guide selection of ideal algal sources. Improved design of macroalgae-derived bio-photocatalysts holds promise for scaling up application in industrial wastewater treatment. Continued multidisciplinary investigations combining nanoscience, biotechnology and environmental engineering are expected to advance this field towards developing nature-inspired solutions for polluted water remediation.

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