



Green Nanotechnology for Arsenic Remediation: Integrating Biosynthesis and Green Chemistry Principles for Sustainable Water Treatment

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ABSTRACT

Arsenic contamination of groundwater represents one of the most significant public health crises of the twenty-first century, affecting an estimated 94-220 million people across at least 70 countries. Conventional remediation technologies, while effective, are often hampered by high operational costs, secondary waste generation, and limited sustainability. This review synthesizes the foundational principles of green chemistry with recent advances in biosynthesized nanomaterials for arsenic decontamination. We examine the mechanistic frameworks underlying green nanoparticle synthesis using plants, microorganisms, and agricultural wastes, with particular emphasis on their application as nano-biosorbents for arsenite and arsenate removal. The integration of bio-based nanomaterials including biochar composites, chitosan derivatives, and metal oxide nanostructures—offers enhanced adsorption capacities, improved selectivity, and inherent environmental compatibility. Critical analysis of current challenges, including scalability, material stability, and ecotoxicological considerations, provides a roadmap for future research directions. This review positions green nanotechnology as a transformative approach to achieving sustainable, affordable, and safe drinking water in arsenic-affected regions worldwide.

KEY WORDS

Green Chemistry, Arsenic Remediation, Biosynthesized Nanoparticles, Nano-biosorbents, Water Treatment, Sustainable Nanotechnology.

INTRODUCTION

1. The Global Arsenic Crisis

Water, the fundamental prerequisite for all recognized forms of life, faces unprecedented contamination challenges in the modern era. While freshwater constitutes merely 2.5% of Earth's total water resources, its uneven distribution 87% in ice caps, 11% in swamps, and only 2% in river systems exacerbates the vulnerability of human populations to water quality degradation. Among the myriad contaminants threatening global water security, arsenic occupies a uniquely perilous position, earning its historical epithet as "The King of Poisons."

Arsenic contamination of groundwater affects at least 106 countries, with recent statistical modeling indicating that 94-220 million individuals are potentially exposed to arsenic concentrations exceeding the World Health Organization's provisional guideline value of 10 µg/L. Bangladesh remains the most devastating case study, where an estimated 35-77 million people face chronic exposure through shallow tubewells, representing one of the largest mass poisonings in human history. The Bengal Delta, encompassing Bangladesh and West Bengal (India), has recorded the most severe arsenic poisoning events, with millions at risk from geogenically contaminated aquifers.

The toxicological profile of arsenic is profoundly species-dependent. Inorganic arsenic predominates in aqueous environments, existing primarily as trivalent arsenite [As(III)] and pentavalent arsenate [As(V)], with As(III) exhibiting significantly greater toxicity and mobility. Chronic exposure manifests through characteristic arsenicosis symptoms hyperpigmentation, keratosis, and ultimately cancers of the skin, lung and bladder while emerging evidence implicates arsenic in cardiovascular disease, neurological impairment, and adverse birth outcomes.

2. Limitations of Conventional Remediation

Traditional arsenic removal technologies coagulation-flocculation, ion exchange, activated alumina adsorption, and membrane processes such as reverse osmosis have demonstrated technical efficacy but face substantial practical limitations. These methods frequently generate toxic metal-laden sludge, require sophisticated infrastructure, consume significant energy, and prove economically prohibitive for resource-constrained communities. The coagulation process, for instance, while capable of achieving regulatory compliance, produces secondary waste streams that pose their own disposal challenges.

The 1993 WHO guideline revision from 50 µg/L to 10 µg/L further strained conventional technologies, as many established treatment systems could not reliably achieve the more stringent standard. This regulatory evolution, combined with the recognition that arsenic disproportionately affects rural populations in developing nations, created an urgent imperative for alternative approaches that balance efficacy with affordability, simplicity, and sustainability.

3. The Emergence of Green Nanotechnology

Green nanotechnology represents the convergence of two transformative paradigms: the size-dependent properties of nanoscale materials and the environmentally benign principles of green chemistry. As articulated by Anastas and Warner, the twelve principles of green chemistry provide a systematic framework for designing chemical products and processes that reduce or eliminate hazardous substances. When applied to nanoscience, these principles guide the development of synthetic methodologies that minimize waste, maximize atom economy, employ renewable feedstocks, and prioritize energy efficiency.

The application of green chemistry to nanotechnology addresses a fundamental tension: while nanomaterials offer unprecedented capabilities for contaminant removal, their synthesis through conventional routes often involves toxic reagents, energy-intensive processes, and the generation of harmful by-products. Traditional nanoparticle synthesis frequently relies on sodium borohydride, hydrazine, or organic solvents substances whose environmental and human health impacts contradict the very goals of remediation.

This review examines the integration of green chemistry principles with nanotechnology for arsenic remediation, focusing on biosynthesized nanomaterials as sustainable alternatives to conventional adsorbents. We explore the mechanistic foundations of green synthesis, evaluate the performance of bio-based nanosorbents, and critically assess the challenges and opportunities for translating laboratory innovations to field-scale applications in arsenic-affected communities.

Principles of Green Chemistry in Nanomaterial Synthesis

1. The Twelve Principles and Their Nanotechnological Applications

The twelve principles of green chemistry, originally formulated to guide sustainable chemical practice, find renewed relevance in the context of nanomaterial synthesis. Table 1 summarizes these principles and their specific application to green nanotechnology.

Table 1: The Twelve Principles of Green Chemistry Applied to Nanotechnology

Principle	Description	Application in Green Nanotechnology
Prevention	Prevent waste rather than treat after creation	Design processes minimizing by-products in nanoparticle synthesis
Atom Economy	Maximize incorporation of all materials into final product	Use methods yielding higher percentages of desired nanoparticles
Less Hazardous Syntheses	Use and generate substances with little or no toxicity	Employ benign solvents and nontoxic precursors
Designing Safer Chemicals	Preserve efficacy while reducing toxicity	Select natural extracts as reducing and capping agents
Safer Solvents and Auxiliaries	Minimize auxiliary substances and ensure safety	Utilize water or bio-based solvents in synthesis
Energy Efficiency	Minimize energy requirements	Conduct synthesis at ambient temperature and pressure
Renewable Feed stocks	Prefer renewable over depleting feed stocks	Utilize biomass and agricultural waste
Reduce Derivatives	Avoid unnecessary derivatization	Streamline synthesis to reduce reagents and waste
Catalysis	Use catalytic rather than stoichiometric reagents	Employ enzymes or biocatalysts for efficient synthesis
Design for Degradation	Design products to degrade into harmless substances	Develop biodegradable nanomaterials
Real-time Analysis	Monitor processes to prevent hazardous formation	Integrate monitoring during synthesis for safety
Inherently Safer Chemistry	Minimize potential for accidents	Design processes reducing explosion or release risks

2. Biological Entities as Green Synthesis Reagents

The cornerstone of green nanomaterial synthesis lies in utilizing biological systems or naturally derived compounds as reducing and stabilizing agents. These bio-resources ranging from whole organisms to isolated biomolecules offer inherent advantages: they operate under mild conditions, require no toxic reagents, and often impart unique surface functionalities through natural capping agents.

Plants have garnered particular attention due to their extensive biodiversity, accessibility, and rich phytochemical profiles. Polyphenols (flavonoids, tannins, phenolic acids), terpenoids, alkaloids, and polysaccharides present in plant extracts possess hydroxyl, carboxyl, and amine groups capable of reducing metal ions and subsequently stabilizing the nascent nanoparticles through steric or electrostatic mechanisms. The hydroxyl groups of polyphenols, for instance, donate electrons for metal ion reduction while their bulky molecular structures prevent nanoparticle agglomeration.

Microorganisms bacteria, fungi, algae offer alternative biosynthetic platforms employing enzymatic reduction mechanisms. Nitrate reductase, NADH-dependent enzymes, and secreted metabolites facilitate nanoparticle formation through intra- or extracellular pathways. Fungi are particularly advantageous for large-scale synthesis due to their high metal tolerance, efficient bioaccumulation, and secretion of substantial quantities of reducing proteins.

Agricultural wastes represent an underexploited but highly promising feedstock for green synthesis. Materials such as rice husk, betel nut husk, fruit peels, and oil cakes embody the renewable feedstocks principle while simultaneously addressing waste management challenges through valorization.

Arsenic Speciation and Remediation Mechanisms

1. Environmental Chemistry of Arsenic

Arsenic's environmental behavior and toxicity are fundamentally governed by its chemical speciation. In aqueous systems, arsenic exists primarily in two oxidation states: arsenite [As(III)] and arsenate [As(V)], with As(III) predominating under reducing conditions (e.g., groundwater aquifers) and As(V) under oxidizing conditions (e.g., surface waters).

The speciation profoundly influences both toxicity and removal efficiency. As(III) exists predominantly as neutral $HfAsOf$ at circumneutral pH, rendering it less amenable to electrostatic adsorption mechanisms that effectively capture anionic As(V) species ($HAsO_4^-$, $H_2AsO_4^{2-}$). This differential behavior necessitates either oxidative pretreatment converting As(III) to As(V) or the development of sorbents with high affinity for both species.

2. Mechanisms of Arsenic Removal by Nanomaterials

Nanomaterials facilitate arsenic removal through multiple complementary mechanisms:

Adsorption represents the primary removal pathway, leveraging the high surface area-to-volume ratios characteristic of nanomaterials. Surface functional groups hydroxyl, carboxyl, amino form complexes with arsenic species through ligand exchange or electrostatic interactions.

Surface complexation involves the formation of inner-sphere or outer-sphere complexes between arsenic oxyanions and metal oxide surfaces. Iron oxides, in particular, exhibit strong affinity for both As(III) and As(V) through bidentate binuclear bridging complexes.

Redox reactions can transform arsenic species, either through oxidation of As(III) to the more readily adsorbable As(V) or through reduction to elemental arsenic or arsine (though the latter is undesirable due to acute toxicity).

Ion exchange mechanisms operate in materials with labile counterions, exchanging harmless ions for arsenate or arsenite.

Photocatalytic degradation, while more relevant to organic pollutants, can facilitate arsenic oxidation through generation of reactive oxygen species.

Biosynthesized Nanomaterials for Arsenic Remediation

1. Biochar-Based Nanocomposites

Biochar, produced through pyrolysis of biomass under oxygen-limited conditions, has emerged as a versatile platform for green nanosorbent development. Its high carbon content, porous structure, and surface functionality provide an excellent substrate for nanoparticle immobilization.

A representative example involves the green synthesis of magnetite-biochar composites (Fe₃O₄@BC) using betel nut husk-derived biochar and iron ore tailings, with *Hibiscus rosa sinensis* leaf extract serving as a natural biosurfactant and reducing agent. This approach embodies multiple green chemistry principles: renewable feedstocks (agricultural waste), waste valorization (mine tailings), and benign synthesis conditions (ambient temperature, aqueous medium).

The optimized composite (Fe₃O₄@BC, Fe:BC ratio 1:2) achieved a BET surface area of 73 m² g⁻¹ with crystalline magnetite nanoparticles uniformly dispersed on the biochar matrix. Batch adsorption experiments demonstrated maximum capacities of 42.74 mg g⁻¹ for As(V) and 27.62 mg g⁻¹ for As(III), with favorable kinetics, minimal interference from coexisting ions, and effective regeneration over three cycles.

2. Chitosan-Based Adsorbents

Chitosan, derived from chitin the second most abundant biopolymer after cellulose exemplifies the renewable feedstocks principle. Its abundant amino and hydroxyl groups provide coordination sites for metal ions while enabling chemical modification to enhance arsenic affinity.

Chitosan composite adsorbents incorporating iron oxides, titanium dioxide, or other metal oxides have demonstrated enhanced arsenic removal through synergistic effects: the biopolymer matrix provides mechanical stability and additional functionality, while the embedded nanoparticles contribute high surface area and specific arsenic binding sites. Magnetic chitosan composites offer the additional advantage of facile separation using external magnetic fields, addressing the recovery challenge that plagues many nanomaterial applications.

3. Metal and Metal Oxide Nanoparticles

Iron-based nanomaterials including zero-valent iron (nZVI), magnetite (Fe₃O₄), maghemite (γ-Fe₂O₃), and goethite (α-FeOOH) have received extensive attention for arsenic remediation due to their high affinity, low cost, and environmental compatibility. Green synthesis routes employing plant extracts (e.g., green tea, *Eucalyptus*, *Azadirachta indica*) produce iron nanoparticles with comparable or superior performance to chemically synthesized analogs.

Zinc oxide nanoparticles synthesized through sustainable routes have demonstrated dual functionality: arsenic adsorption coupled with photocatalytic oxidation of As(III) to As(V) under solar irradiation. The green synthesis approach, utilizing plant phytochemicals as reducing and capping agents, yields nanoparticles with enhanced biocompatibility and reduced ecotoxicity compared to conventionally produced ZnO.

Titanium dioxide nanomaterials offer photocatalytic capabilities that oxidize As(III) to the more readily adsorbable As(V) while simultaneously degrading organic co-contaminants. Green synthesis using algal or plant extracts produces TiO₂ nanoparticles with tailored crystal phases (anatase vs. rutile) and surface properties optimized for arsenic removal.

4. Comparative Performance Analysis

Table 2: Biosynthesized Nanomaterials for Arsenic Removal

Nanomaterial Type	Biosynthesis Agent	Target Species	Adsorption Capacity (mg g ⁻¹)	Key Features
Fe ₃ O ₄ @BC composite	<i>Hibiscus</i> leaf extract	As(V), As(III)	42.74 (AsV), 27.62 (AsIII)	Waste-derived, regenerable
Chitosan-Fe composite	Crustacean shells	As(V)	35.2	Biodegradable, magnetic separation
Iron oxide NPs	Green tea extract	As(III), As(V)	28.5	Ambient synthesis, stable
ZnO nanoparticles	Plant extracts	As(III)	23.8	Photocatalytic oxidation
Zero-valent iron	<i>Eucalyptus</i> extract	As(III)	31.2	High reactivity

Mechanistic Insights into Biosorption

1. Equilibrium and Kinetics

Biosorption involves the partitioning of dissolved arsenic species between aqueous solution and solid biosorbent phases until equilibrium is established. The process follows characteristic patterns describable by adsorption isotherms (Langmuir, Freundlich, Temkin) and kinetic models (pseudo-first-order, pseudo-second-order, intraparticle diffusion).

The Langmuir model, assuming monolayer adsorption on homogeneous surfaces with finite identical sites, frequently provides excellent fit for arsenic biosorption data, yielding maximum capacity values and affinity constants. The Freundlich model, describing multilayer adsorption on heterogeneous surfaces, often better represents biosorbents with diverse surface functionalities.

Kinetic studies typically reveal rapid initial uptake (minutes to hours) followed by slower approach to equilibrium, suggesting involvement of multiple rate-limiting steps including film diffusion, intraparticle diffusion, and surface complexation reactions.

2. Factors Influencing Biosorption Efficiency

pH exerts dominant influence on arsenic biosorption by governing both arsenic speciation and surface charge of biosorbents. As(V) removal typically maximizes under acidic to neutral conditions (pH 3-7) where anionic H₂AsO₄⁻ predominates and positively charged biosorbent surfaces facilitate electrostatic attraction. As(III) removal often shows broader pH optima due to the neutral HfAsOf species, with removal mediated primarily through inner-sphere complexation rather than electrostatics.

Biosorbent dosage affects removal efficiency through availability of binding sites, with increasing dosage typically enhancing percentage removal while reducing capacity due to site underutilization.

Competing ions particularly phosphate, silicate, and carbonate can significantly reduce arsenic removal through competition for binding sites or modification of surface charge. The extent of interference depends on both anion concentration and affinity for the biosorbent surface.

Temperature influences both equilibrium capacity (typically endothermic adsorption processes show enhanced capacity at elevated temperatures) and kinetic rates.

3. Regeneration and Reuse

Biosorbent regeneration is essential for economic viability and waste minimization. Effective desorption of arsenic enables both biosorbent reuse and, potentially, arsenic recovery. Dilute NaOH solutions effectively elute adsorbed arsenic through pH-induced reversal of surface complexation, while maintaining biosorbent integrity for subsequent cycles.

Successful regeneration over multiple cycles (typically 3-5) has been demonstrated for various biosorbents, though gradual capacity decline necessitates eventual replacement. The spent biosorbent, if non-toxic and biodegradable, may be disposed of through incineration with significant volume reduction, or potentially used in controlled land application if arsenic content meets regulatory standards.

Challenges and Future Perspectives

1. **Scalability and Reproducibility:** Despite promising laboratory results, translation of green nanosorbents to field applications faces substantial hurdles. Batch synthesis using plant extracts exhibits inherent variability due to seasonal, geographical, and genotypic differences in phytochemical composition. This variability complicates quality control and reproducible performance a critical requirement for regulatory approval and user acceptance.

Scale-up from milligram laboratory syntheses to kilogram quantities required for community-scale treatment systems demands process engineering innovations. Continuous flow reactors, standardized biomass processing, and formulation into practical deployment formats (cartridges, packed columns, impregnated filters) remain underdeveloped.

2. **Material Stability and Performance in Complex Matrices:** Real groundwater matrices contain multiple solutes dissolved organic matter, competing anions, natural organic matter that can compromise laboratory-optimized performance. Long-term stability under field conditions, including resistance to microbial degradation, oxidative transformation, and mechanical attrition, requires systematic investigation.
3. **Ecotoxicological Considerations:** Paradoxically, nanomaterials designed for environmental remediation may themselves pose environmental risks if released during production, application, or disposal. The unique properties enabling efficient contaminant removal high reactivity, small size, surface functionality also govern their environmental fate and potential toxicity.

Green synthesis approaches, employing natural capping agents and biodegradable matrices, may reduce but not eliminate ecotoxicological concerns. Comprehensive life-cycle assessment, encompassing raw material extraction, synthesis, application, and end-of-life scenarios, remains essential for establishing true environmental superiority over conventional alternatives.

4. **Integration with Existing Water Infrastructure:** Successful deployment requires compatibility with existing water supply systems and user practices. Point-of-use devices, community-scale treatment plants, and piped water systems impose different constraints on material format, flow rates, and maintenance requirements. Integration with solar-powered systems, smart sensing technologies for breakthrough detection, and remote monitoring capabilities represent promising directions for enhancing practical utility.
5. **Future Research Directions:** Mechanistic understanding at molecular and atomic scales through advanced spectroscopic techniques (XAS, EXAFS) and computational modeling will enable rational design of biosorbents with enhanced selectivity and capacity.

Hybrid systems combining biosorption with membrane filtration, photocatalytic oxidation, or electrochemical processes may overcome individual limitations while leveraging complementary strengths.

Waste valorization converting agricultural, industrial, or municipal wastes into value-added biosorbents embodies circular economy principles while addressing multiple environmental challenges simultaneously.

Field validation through carefully designed pilot studies in arsenic-affected communities, incorporating socio-economic dimensions alongside technical performance, will bridge the gap between laboratory innovation and real-world impact.

CONCLUSIONS

Green nanotechnology, integrating the foundational principles of green chemistry with the unique capabilities of nanoscale materials, offers transformative potential for addressing the global arsenic crisis. Biosynthesized nanomaterials derived from plants, microorganisms, and agricultural wastes provide arsenic removal capacities comparable to conventional adsorbents while embodying environmental compatibility, cost-effectiveness, and sustainability.

The mechanistic diversity of biosorption encompassing surface complexation, electrostatic interactions, redox transformations, and ion exchange enables effective removal of both arsenite and arsenate across varied water matrices. Biochar composites, chitosan derivatives, and biosynthesized metal oxides have demonstrated particular promise, with adsorption capacities reaching 40-50 mg g⁻¹ and regeneration capabilities supporting multiple treatment cycles.

Yet significant challenges remain. Scalable, reproducible synthesis; long-term stability in complex environmental matrices; comprehensive ecotoxicological assessment; and integration with existing water infrastructure demand sustained interdisciplinary effort. The translation of laboratory innovations to field-scale applications in resource-constrained communities where the arsenic burden falls heaviest requires not only technical optimization but also attention to affordability, cultural acceptability, and institutional capacity.

The green synthesis paradigm, by aligning nanomaterial production with environmental stewardship, offers a path toward remediation technologies that heal rather than harm solutions that address today's contamination without creating tomorrow's problems. As global arsenic contamination continues to threaten millions of lives, the imperative for such sustainable approaches has never been more urgent.

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