

REVIEW

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Green synthesis trends and potential applications of bimetallic nanoparticles towards the sustainable development goals 2030

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The world faces threats that the United Nations has classified into 17 categories with different objectives as solutions for each challenge that are enclosed in the Sustainable Development Goals (SDGs). These actions involved the widespread use of science and technology as pathways to ensure their implementation. In this regard, sustainability science seeks the research community's contribution to addressing sustainable development challenges. Specifically, nanotechnology has been recognized as a key tool to provide disruptive and effective strategies to reach the SDGs. This review proposes the application of bimetallic nanoparticle substances capable of providing possible solutions to achieve target SDG 3: good health and well-being, SDG 6: clean water and sanitation, and SDG 12: responsible consumption and production. Furthermore, the term green nanotechnology is introduced in each section to exemplify how green synthesized bimetallic nanoparticles have been used to resolve each target SDG. This review also outlines the current scenario regarding the utilization of metallic nanomaterials in the market, together with the upscaling challenges and the lack of understanding of the long-term effects and hazards to the environment regarding bimetallic nanoparticles.

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1 Introduction

In September 2015, 193 states adopted the United Nations (UN) 2030 Agenda for Sustainable Development.¹ This agenda is a comprehensive policy blueprint that aims for all nations to be economically prosperous, socially inclusive, environmentally sustainable, and well-governed by 2030. To achieve these objectives, 17 Sustainable Development Goals (SDGs) were established and divided into four main pillars: social, economic, environmental, and governance.² The social pillar comprises the first seven goals of the agenda, focusing on tackling poverty, bad nutrition, a deficit in ensuring healthy lives, deficiency in education systems, gender inequality, and lack of clean water and sanitation and aims to provide access to sustainable energy. The economic pillar encompasses goals 8 to 12, which involve decent work, economic growth, innovation, infrastructure, income inequalities, sustainable cities, and responsible consumption and production. Goals 13 to 15 comprise the environmental pillar and target how to take care of our planet in terms of climate, life underwater, and land, respectively. Finally, the governance pillar comprises goals 16

and 17 related to promoting peace and strong institutions and partnerships, respectively, aligned with the previous SDGs.

To accomplish the SDGs, the UN established the need to mobilize resources from a variety of financial and technological sources, among other means of implementation.³ For this reason, the UN Department of Economic and Social Affairs published the *Global Sustainable Development Report (GSDR) 2019: The Future is Now – Science for Achieving Sustainable Development*.⁴ In this report, the scientific and technological contributions are defined as the pathway to sustainability transformation and technological innovation to shifting away from the creation of business-as-usual together with the action of scaling up applications of scientific knowledge.⁴ It also identifies science, technology, and innovation as central tools for SDG implementation and explains how technology is usually already available.⁴ Finally, it recommends prioritizing the strategic deployment of novel technologies to minimize the issues to achieve the goals.⁴

Over the past few years, reports have shown how nanostructured materials (or nanomaterials) can make a difference in applications in health and well-being, water treatment and sanitation, and responsible consumption and production of resources. An example is how metal-based nanostructures have been implemented in environmental remediation, electronics, gene therapy, and drug delivery solutions; this is because of their remarkable characteristics, like their higher surface area, surface energy, and chemical reactivity concerning macroscopic materials.⁵ Specifically, bimetallic nanoparticles (BMNPs) have

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In general, BMNPs are synthesized by two general approaches, *i.e.*, the top-down and the bottom-up methods.⁵⁰ The first approach is usually achieved through physical methods, and the second by chemical and biological methods.⁵⁰ In the top-down approach, the bulk material is fragmented into nano-sized structures in the presence or absence of a catalyst.⁵¹ In contrast, the bottom-up methods include extending atomic and molecular precursors to the nanoscale.⁵² The chemical synthesis by co-reduction of two ionic species, a bottom-up method, is one of the most commonly used chemical strategies to synthesize noble metal-based BMNPs, while transition metal-based BMNPs are

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Table 1 Traditionally synthesized bimetallic nanoparticles (T-BMNPs) with potential applications towards SDGs 3, 6, and 12. M1/M2, M1@M2, and M1M2 notations denote alloy, core-shell, and BMNPs with undefined structures, respectively. NS: not specified

NPs	Synthesis method	Size (nm)	Morphology	Application	SDG	References
Pd/Cu	Surfactant-directed aqueous method	3	Nanowires	Quantitative determination of organophosphate pesticides in vegetables and fruits	3	22
Fe/Pd	NaBH ₄ reduction	155	NS	Degradation of soil-sorbed trichloroethylene	3	41
Fe/Ag	NaBH ₄ reduction	80	Spherical	Dechlorination and oxidative degradation of 4-chlorophenol	3	43
FeCu	NaBH ₄ reduction	100	NS	Debromination of 2,2',4,4'-tetrabromodiphenyl ether (BDE-47)	3	26
FeNi						
FePd						
FeAg						
FePt						
FeAu						
Fe/Pd	NaBH ₄ reduction	3–6	Spherical	Dechlorination of excess trichloroethene before aquifer spread	3	42
Fe/Ni						
Fe/Cu						
Fe/Ag						
Ag/Ni	NaBH ₄ reduction	20–25	Quasi-spherical	Reusable catalytic system for efficient reduction of <i>p</i> -nitrophenol to <i>p</i> -aminophenol	3, 12	45
Ag/Cu	NaBH ₄ reduction	NS	NS	Reduction of 4-nitrophenol to 4-amino-phenol	3, 6	44
CuAg	NaBH ₄ reduction	59	Spherical	Reduction of nitrophenols and organic dyes	3, 6	65
CuNi		68				
Cu@Fe	NaBH ₄ reduction	70	Spherical irregular	Removal of cesium from contaminated water	6, 12	66
Ag/Au	NS	9.6	Spherical	Detection of Cr(VI) ions in water	6	30
Fe/Ni	NaBH ₄ reduction	100	Spherical	Antibiotic tetracycline removal	6, 3	67
Ag/Cu	NS	<100	NS	Packaging material for food preservation	12	68
Ag/Cu	NS	<100	NS	Active chicken meat packaging	12	69
Ag/Cu	NS	<100	NS	Active chicken meat packaging	12	70
CuNi	NaBH ₄ reduction	3.9	Spherical	Hydrogen evolution from ammonia borane hydrolysis	12	71

synthesized using thermal decomposition due to the inability of the NPs to crystallize at room temperature.²⁹ The size, shape, and stability of NPs can be adjusted by controlling the thermodynamic and kinetic conditions in the synthesis.⁵³

Green nanotechnology overcomes the main restrictions of traditional physicochemical approaches, integrating the principles of green chemistry to create eco-friendly and safe synthesis processes using biological sources⁴⁹ and to incorporate large-scale manufacturing and industrial use of nanomaterials with less environmental degradation and potential risks of human health hazards.⁵⁵ BMNPs that can be synthesized in an environmentally friendly fashion following a green chemistry approach that incorporates at least three factors: the use of nontoxic capping agents, less hazardous reducing agents, and the selection of environmentally benign solvents.⁵⁶ Thus, applying these green chemistry principles to the synthesis of nanomaterials made possible the emergence of the field of green nanotechnology.⁴⁹ In this sense, the introduction of green nanotechnology to sustainability science in the synthesis and the application of BMNPs have shown effective results towards soil and water contamination as well as an effective antibacterial activity against pathogen bacterial strains in the health sector and food industry.

Bacteria, fungi, plants, algae, and organic waste such as fruit peels and the biomolecules that compose them are biological sources generally used as reducing and stabilizing agents⁵⁷ (Fig. 2). In both bacteria and fungi-mediated synthesis, metallic salts can be reduced in an intracellular or extracellular environment.⁶ Bacteria-mediated green synthesis of BMNPs can generate zero-valence metallic structures, which can be implemented in biomedical and technological applications.⁶ However, fungi-mediated syntheses are preferable to bacteria-mediated ones since the first ones can be used in scaled-up, faster processes.^{58,59} The plant-mediated syntheses of metallic NPs are some of the most commonly used processes. Usually, these processes start with preparing a liquid extract of the plant, which results in a fine powder, which must then be washed and dried before the synthesis.⁶⁰ These reactions are favorable since plant leaves comprise several functional groups and phytochemicals acting as stabilizing and reducing agents.⁶¹ Polyethylene glycol⁶² and starch⁶³ are some of the commonly used materials to achieve a biomolecule-mediated synthesis of metallic NPs. Lastly, waste material-mediated synthesis is one of the less explored types of synthesis.⁶ Nevertheless, its implementation in scale-up processes can highlight circular-economy-based models. Generally, the type of waste used





Fig. 2 General methodology for the synthesis of BMNPs via green chemistry methods. Reproduced from ref. 54 with permission from American Chemical Society, copyright 2010.

comes from agricultural wastes, such as fruit peels and organic waste materials. Compared to traditional (also known as “physicochemical”) methods, a remarkable advantage is that green synthesis involves fewer reactants, since, in most cases, reducing and stabilizing agents are the same substance.⁶¹ Furthermore, green nanotechnology presents the advantage of energy efficiency and cost-effectiveness as the synthesis carried out following its principles is low in energy consumption and, consequently, low cost. In addition, they are clean and easy and show a decrease in the production of greenhouse gasses and wastes, as well as a minimal consumption of non-renewable raw materials.^{55,64} BMNPs listed in Table 2 are examples of nanoparticles synthesized by green chemistry methodologies.

The following sections will describe the applications of traditionally and green-synthesized BMNPs (T-BMNPs and G-BMNPs, respectively) towards SDGs 3, 6, and 12. In some cases, one bimetallic nanosystem can be classified into two or more SDGs.

4 Case studies of bimetallic nanoparticles in relation to SDG 3: good health and well-being

SDG 3 prioritizes the assurance of healthy lives and promotes well-being for all ages.⁹⁹ Within the specific targets to achieve this aim, targets 3.3 and 3.9 can be approached by applying BMNPs as a possible solution. On the one hand, target 3.3 aims to end the epidemics of AIDS, tuberculosis, malaria, and neglected tropical diseases, as well as combating hepatitis, water-borne diseases, and other communicable diseases by 2030. Target 3.9, on the other hand, points to substantially

reducing the number of deaths and illnesses produced by hazardous chemicals and contaminants in polluted air, water, and soil.¹⁰⁰ Target 3.9 is important as chemicals used in everyday life goods can greatly affect humans and the environment.¹⁰¹ Fig. 3 summarizes the latest applications of T-BMNPs and G-BMNPs as potential tools for environmental remediation and identification of hazardous chemicals, which will be discussed in the following sections.

4.1 Chemical agent degradation as a pathway for avoiding diseases

Pesticides are one of the types of hazardous chemical agents found in soil. According to recent studies, pesticide pollution is widespread, and its toxicity is a major global health problem.^{102,103} Three million poisoning cases are reported yearly, and between 220 000 and 250 000 deaths occur due to pesticides, where the most common are organophosphates (OPs), specifically malathion.¹⁰³ Given these statistics and the public health concern of pesticide residues in food,^{104–106} Song *et al.*²² developed a highly sensitive, simple, and cost-effective acetylcholinesterase (AChE) electrochemical biosensor for the quantitative determination of malathion in vegetables and fruits based on palladium–copper nanowires (Pd/Cu NWs). The morphology and size of the loaded Pd/Cu NWs on a glass carbon electrode (GCE) showed a high-quality intertwining nanowire morphology with no byproducts such as nanoparticles. The effect of shape control can be attributed to the surfactant-directed aqueous method used to synthesize the bimetallic NWs, as similar shape control results have been reported using this method.¹⁰⁷ Furthermore, according to the results of electrochemical impedance spectroscopy, Pd/Cu NWs could either enhance the conductivity of the electrode interface or facilitate

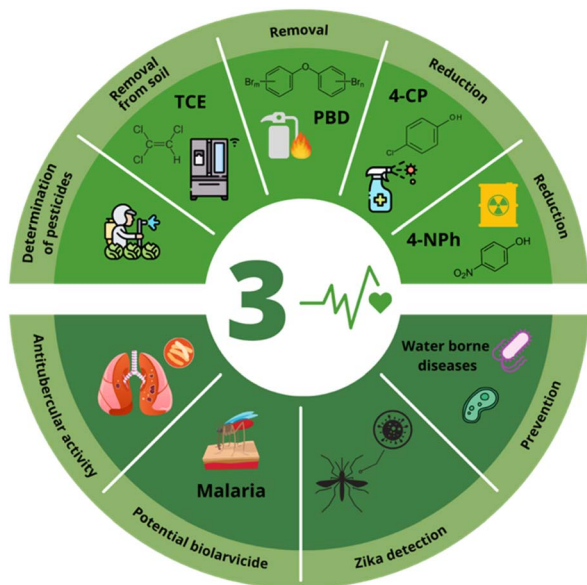


Table 2 Green synthesized bimetallic nanoparticles (BMNPs) with potential applications towards SDGs 3, 6, and 12. M1/M2, M1@M2, and M1M2 notations denote alloy, core-shell, and undefined BMNPs, respectively

NPs	Synthesis method	Reducing agent	Stabilizing agent	Size (nm) and morphology	Application	SDG	References
Au/Ag	Bioreduction	<i>Barleria prionitis</i> <i>Plumbago zeylanica</i> <i>Syzygium cumini</i>	<i>Barleria prionitis</i> <i>Plumbago zeylanica</i> <i>Syzygium cumini</i>	10–70 not defined 90 hexagonal 10–20 spherical	Antitubercular activity	3	72
Fe/Pd	Bioreduction	Green tea	Green tea	20–100 spherical	Trichloroethylene degradation Congo red dye degradation	3	73
Ir@Cu	Chemical reduction	Ethylene glycol	PVP	1.57 spherical	Reduction of the substance 4-nitrophenol	3	74
Fe@Ag	Bioreduction	<i>Salvia officinalis</i>	<i>Salvia officinalis</i>	48 spherical	Zika virus RNA detection	3	24
Au/Ag	Bioreduction	Tannic acid	Tri-sodium citrate	2.9 ± 1.1 spherical		3	75
Au@Ag	Bioreduction			4.4 ± 0.6 spherical			
PdCo	Bioreduction	<i>Cinnamomum verum</i>	<i>Cinnamomum verum</i>	2.4 spherical	Dopamine detection and antibacterial activity against <i>S. aureus</i> and <i>E. coli</i>	3	76
AgAu	Bioreduction	<i>Eichhorniacrassipes</i>	<i>Eichhorniacrassipes</i>	0.31–1.08 spherical and cubic	Biosorption of heavy metals (Pb, Zn, Cu, and Mn)	3, 6	77
FeNi	Bioreduction	<i>Pomegranate peel</i>	CMC	129 ± 7 spherical	Antibiotic tetracycline removal	3, 6, 12	78
Fe/Cu	Bioreduction	<i>Pomegranate peel</i>	CMC	60 spherical	Antibiotic tetracycline removal	3, 6, 12	79
Fe@Pd	Bioreduction	<i>Pomegranate peel</i>	CMC	40 spherical	Antibiotic tetracycline removal	3, 6, 12	80
Ag/Cu	Bioreduction	<i>Carica papaya</i>	<i>Carica papaya</i>	150 star-like	Degradation of chlorpyrifos pesticide in water	6	81
Fe/Pd	Bioreduction	<i>Eucalyptus</i>	<i>Eucalyptus</i>	30–60 spherical	Removal of arsenic from natural drinking water	6	82
Cu/Zn	Bioreduction	<i>Hibiscus rosa sinensis</i>	<i>Hibiscus rosa sinensis</i>	Not defined	Antimicrobial activity in hydrocolloid films	12	83
Au/Pt	Microwave-assisted reduction method	Trisodium citrate	—	20–40 spherical, triangle, ellipsoidal	Antibacterial activity against <i>E. coli</i> , <i>S. typhi</i> , <i>Klebsiella</i> , <i>E. coli</i> and <i>E. faecalis</i>	12, 3	84
Ag/Au	Bioreduction	<i>Plumbago zeylanica</i>	<i>Plumbago zeylanica</i>	93 hexagonal and spherical	Antibiofilm activity against <i>E. coli</i> , <i>A. baumannii</i> , <i>S. aureus</i> , and a mixed culture of <i>A. baumannii</i> and <i>S. aureus</i>	12, 3	85
Ag/Au	Bioreduction	<i>Annona squamosa</i> L.	<i>Annona squamosa</i> L.	30–50 diverse	Antibacterial activity against <i>B. subtilis</i> , <i>S. aureus</i> , <i>E. coli</i> and <i>S. typh</i>	12, 3	86
Cu@Pt	Bioreduction	<i>Agrimoniae herba</i>	<i>Agrimoniae herba</i>	30 spherical	Antibacterial activity against <i>E. coli</i> , <i>S. aureus</i> , and <i>P. aeruginosa</i>	12, 3	87
Cu/Au	Bioreduction	<i>Aspergillus niger</i>	—	23–199 triangular	Antimicrobial activity <i>E. coli</i> , <i>K. pneumonia</i> , <i>P. aeruginosa</i> , <i>P. vulgari</i> , <i>S. aureus</i> , <i>E. faecalis</i> , and fungal strain of <i>A. niger</i> and <i>C. albicans</i>	12, 3	88
Fe/Mn	Bioreduction	Auxin complex (indole-3-acetic)	Auxin complex (indole-3-acetic)	250–300 spherical	Plant bio-fertilizer	12	89
Pt@Ag	Bioreduction	<i>Hibiscus sabdaridha</i>	<i>Hibiscus sabdaridha</i>	5–6 spherical	Hydrogen evolution and antibacterial activity	12, 3	90
Pd–Ag	Bioreduction	<i>Nigella Sativa</i>	<i>Nigella Sativa</i>	6–7 spherical	Hydrogen production, antibacterial and anticancer activities	12, 3	91
Pt–Ag	Bioreduction	<i>Nigella Sativa</i>	<i>Nigella Sativa</i>	5–6 spherical	Hydrogen production, antibacterial activity and photodegradation of azo dyes	12, 6, 3	92



NPs	Synthesis method	Reducing agent	Stabilizing agent	Size (nm) and morphology	Application	SDG	References
PdPt	Bioreduction	<i>Nigella Sativa</i>	<i>Nigella Sativa</i>	10–25 spherical	Hydrogen production, antibacterial and anticancer activities	12, 3	93
Pd–Ag	Bioreduction	Stevia extract	Stevia extract	5–15 spherical	Hydrogen production and antibacterial activity	12, 3	94
Fe–Cr	Coprecipitation with simulated industrial effluents	—	—	20 spherical	Ferromagnetic activity for smart material applications	12	95
Ag/Cu	Chemical reduction	Ascorbic acid	Chitosan	50–80 spherical	Antibacterial activity against <i>E. Coli</i> , <i>P. aeruginosa</i> and <i>S. typhi</i>	12	96
Au–Pt	Bioreduction	<i>Pleurotus florida</i>	<i>Pleurotus florida</i>	16 icosahedral	Agro-waste management and anticancer activity	12, 3	97
Fe–Zn	Bioreduction	<i>Citrus limon</i> (L.) <i>Burm. f.</i> wastes	<i>Citrus limon</i> (L.) <i>Burm. f.</i> wastes	126 spherical	Decolorization of reactive-red 2 (RR2) dye	12, 6	98



electron transfer, as was also reported by Hasanjani and Zarei.¹⁰⁸ Additionally, bimetallic NWs increased the electroactive surface area compared to the bare GCE. Finally, the biosensor indicated an acceptable reproducibility, and its limit of detection (LOD) was estimated to be 1.5 parts per trillion (ppt). This result was lower than the European maximum residue levels of malathion for courgettes, carrots, lettuces, and oranges as well as those obtained by Liquid Chromatography–Mass Spectroscopy method, in which pesticides at concentrations lower than 1.4 part per billion (ppb) cannot be detected.²² These results show that BMNP-based biosensors can potentially be used to comply with pesticide regulations protecting food consumers.

One of the most persistent synthetic phenols used as pesticides or herbicides is a group of chemicals produced by

electrophilic halogenation of phenol with chlorine called chlorophenols.¹⁰⁹ Barreto-Rodrigues *et al.*⁴³ prepared BMNPs of Fe and Ag immobilized in calcium alginate beads (Fe/Ag-Alg) to test dechlorination and oxidative degradation of 4-chlorophenol (4-CP). Batchwise experiments showed that Fe/Ag-Alg completely removed 4-CP under acidic conditions (pH 3). At low pH, BMNPs displayed a low removal of total organic carbon (TOC), which can be explained as the dechlorination process does not allow further degradation, and the reaction products, such as phenols, still need additional treatment. It also presented low Fe leaching compared to bare Fe/Ag. This demonstrates the stability and effective immobilization of Fe/Ag NPs in alginate. Under the same conditions, H₂O₂ was added after dechlorination by Fe/Ag-Alg to increase the TOC removal. Results indicated that adding H₂O₂ completely removed 4-CP and TOC with a relatively low Fe leaching. The results obtained are important as TOC reflects water quality, and according to trials, reducing TOC decreases the formation of carcinogenic compounds such as trihalomethanes (THM).¹¹⁰

Tuberculosis (TB) is caused by *Mycobacterium tuberculosis* and affects millions of people every year.¹¹¹ The resistance of *M. tuberculosis* strains to the four first-line drugs, such as rifampicin, isoniazid, pyrazinamide, and ethambutol, has become a worldwide problem.¹¹¹ Singh and coworkers reported the synthesis of Au/Ag NPs mediated by three different medicinal plants as reducing agents (*B. prionitis*, *P. zeylanica*, and *S. cumini*) and the evaluation of their antitubercular activity.⁷² Results from preliminary screening assays showed that the three bimetallic nanosystems inhibited mycobacterial growth by 90%, indicating that BMNPs have a profound mycobactericidal potency. Moreover, *in vivo* and *ex vivo* macrophage infection assays showed that *S. cumini*-Au/Ag NPs effectively inhibit both active and dormant mycobacteria at a low minimum inhibitory concentration (MIC) and had a greater specificity towards mycobacteria in contrast with the host cells. Although the result did not show rifampicin's high selectivity, Au/Ag NPs can be used as an attractive strategy to overcome rifampicin drug resistance and treat tuberculosis.⁷² Gopinath

*et al.*¹¹² also synthesized Au/Ag NPs using *Terminalia arjuna* as a reducing agent to control the malaria vector *Anopheles stephensi*. In the larvicidal bioassay, Au/Ag NPs showed a higher susceptibility rate in contrast with individual Ag or Au NPs. Therefore, it was concluded that phyto-mediated synthesized metal NPs could potentially be used to control malaria transmission, diminishing long-term undesirable effects produced by synthetic insecticides.¹¹²

4.2 Targeting human exposure to soil pollutants

Retaking the soil pollution approach, trichloroethylene (TCE) is another health-alarming soil chemical pollutant. TCE is an organic compound with low water solubility and one of the most widely chlorinated hydrocarbons used in industry.¹¹³ From its whole annual production, it is estimated that 83.60% of the TCE's annual production volume is used as an intermediate in the manufacture of hydrofluorocarbon HFC-134a,¹¹⁴ which is primarily used as a refrigerant for domestic refrigeration and automobile air conditioners.^{114,115} TCE's low solubility and high absorbance by soil organic matter (SOM) may produce a water/soil distribution, leading to environmental contamination with human health implications.¹¹⁴ According to Dumas, chronic TCE exposure through industrial operations or contaminated drinking water can affect vital organs such as the liver and kidneys.¹¹³ Dechlorination of TCE using BMNPs has been proposed as a remediation strategy to diminish its environmental impact on contaminated soil and groundwater.^{41,116,117} FePd NPs stabilized with carboxymethyl cellulose (CMC) have been used to perform TCE *in situ* dechlorination.^{41,116} Zhang *et al.* studied the reactivity of BMNPs, testing TCE degradation by spherical FePd NPs stabilized with 0.2% CMC.⁴¹ Degradation results showed that after 40 min BMNPs led to the destruction of nearly all the initial TCE. One of the critical aspects during the reaction of TCE degradation is the production of toxic intermediates. Nevertheless, the mass balance of the presented FePd NPs indicated rapid and nearly complete TCE dechlorination without the presence of vinyl chloride and dichloroethenes as by-products. Besides, CMC monodentate interaction with FePd NPs suppressed the growth of nanoparticles, maintaining a higher surface area, provided stability against aggregation in water, and allowed NPs to pass through and be thoroughly dispersed in the soil bed, in contrast to non-stabilized FePd NPs, which were retained on the top of the soil bed. These results were obtained in the presence of a surfactant, and the evaluation of its activity was held in two different soil matrices, and they were comparable with the ones obtained by He *et al.*⁴²

The TCE degradation properties of BMNPs were also tested by Smuleac *et al.*,⁷³ following a green synthesis method. In this study, bimetallic Fe/Pd NPs were immobilized in a polyacrylic acid, PAA-coated polyvinylidene fluoride membrane. First, Fe NPs were loaded into the polymeric membrane, immersing it into an FeCl₂ solution, which functioned as the precursor salt. The reducing and capping agent utilized for the synthesis was a green tea extract, which has proved to contain polyphenols in a concentration that makes it suitable for the synthesis. After

the Fe²⁺ modified membrane was prepared, it was immersed in the green tea extract solution. The third and last step after synthesizing the Fe NPs within the membrane was another process of immersion, this time in a K₂PdCl₄ solution, for the loading of Pd into the Fe NPs. For the TCE degradation performance study, small pieces of the membrane were cut and immersed into a TCE solution. Studies were carried out first with a membrane that contained only Fe NPs, and then with the BMNP membrane. The Fe/Pd-based system showed an increase of its k_{SA} from 0.005 to 0.008 L m⁻², reducing the concentration of TCE to almost 35% of the original concentration, while the solely Fe loaded membrane reduced it to around 60%. The study demonstrated how the bimetalization of NPs can lead to higher degradation activity of TCE. These results are important due to the increasing demand for industrial refrigeration systems across the fast-moving consumer goods sector¹¹⁸ and, as a consequence, the use of TCE.

4.3 Treating industrial wastes

Other persistent organic pollutants in the environment are polybrominated diphenyl ethers (PBDEs). PBDEs are toxic halogenated aromatic compounds used in different industries that generally lack safety precautions in their management and disposal.¹¹⁹ In addition, its high potential for long-range environmental transport can easily produce bio-accumulation in food chains, generating critical adverse health effects in different sectors of the population.¹²⁰ Wang *et al.*²⁶ studied the debromination mechanism of 2,2',4,4'-tetrabromodiphenyl ether (BDE-47) by six different bimetallic systems Fe/M (M: Cu, Ni, Pd, Ag, Pt, and Au). The first important factor they identified was that all the metal additive powders were unreactive toward BDE-47 in the absence of Fe. Therefore, the strong reductive capability of Fe is the primary source of these systems' enhanced activity. Second, the debromination rates of BDE-47 in all the bimetallic systems at the same concentration of metal additive decreased in the following order: Fe/Pd > Fe/Ag > Fe/Cu > Fe/Ni > Fe/Au > Fe/Pt $\approx$ *n*-ZVI. Third and last, debromination pathways are different for each system. Fe/Ni, Fe/Pd, and Fe/Pt systems have an H-transfer dominant mechanism and generate BDE-17 as the final product. The Fe/Ag system follows an electron-transfer dominant mechanism with BDE-28 as the final product. Finally, Fe/Cu and Fe/Au systems presented a combination of the two mechanisms generating BDE-17 and BDE-28 as final products. This work contributed to understanding debromination and proposed a solution for PBDE pollution to promote well-being. Important to mention is that pre- or post-natal exposure to PBDEs may cause long-lasting behavioral abnormalities, particularly in motor activity and cognition.¹²¹ Apart from PBDEs, synthetic phenols are persistent chemicals that show toxicity.¹²⁰ An example of this is the study performed by Yang *et al.*,¹²² who found that the exposure of zebrafish to synthetic phenolic antioxidants in a range concentration between 5 and 200 μ M produced morphological deformities and abnormal pigmentation in their bodies.

The reduction of nitroaromatic (NA) compounds, another type of industrial waste pollutant, can be performed by BMNPs. For



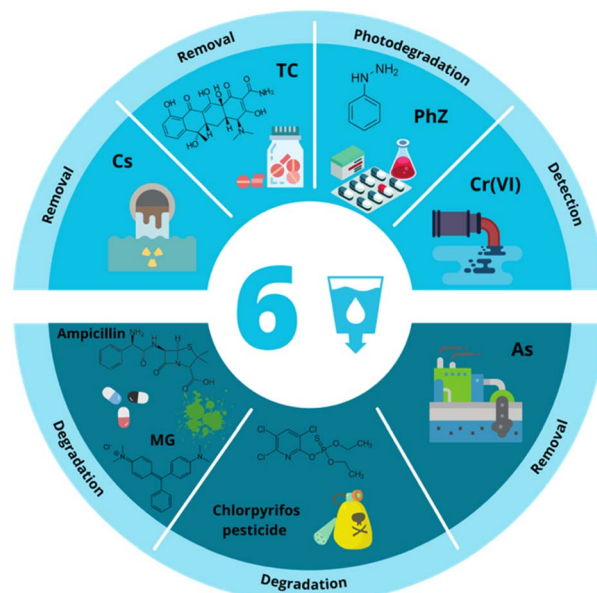
Congo red (CR) dye was also successfully oxidized by utilizing Ir@Cu BMNPs in the work of Pooja and Anjali Goel.⁷⁴ Core-shell Ir@Cu BMNPs were synthesized using ethylene glycol (EG) as an effective reducing agent, together with

Despite the activity of BMNPs reducing 4-NPh to 4-aminophenol (4-Aph), the use of NaBH_4 as a reducing agent in their synthesis method introduces an environmental hazard,⁴⁶ which can be overcome by implementing green methods to synthesize bimetallic systems for this application. Malik *et al.*²⁴ reported green synthesized Fe@Ag nanoparticles using *Salvia officinalis* as a reducing and capping agent to reduce 4-NPh to 4-Aph. FTIR spectroscopic analysis showed that the surface of BMNPs is predominantly composed of polyphenols. These compounds protected zero-valent Fe from oxidation and prevented the agglomeration and aggregation of the NPs, maintaining the catalytic properties of Fe@Ag NPs. The percentage of reduction of 4-NPh to 4-Aph was close to the results shown when T-BMNP Ag/Cu NPs were used,⁴⁴ as well as their reusability. These characteristics made Fe@Ag NPs an effective green catalyst to reduce 4-NPh, one of the 114 more noxious organic pollutants according to the Environmental Protection Agency (EPA), to 4-Aph, which is an intermediate in the synthesis of paracetamol and has important applicability in the textile, leather, and alimentary industries.⁴⁴ Malik *et al.* work is of interest due to an effective reduction of 4-NPh, producing an industrially valued chemical and promoting well-being as localized 4-NPh transformation would eradicate the symptoms generated by its chronic exposure and its genotoxicity.¹²⁴ Continuing this health approach, G-BMNPs have been reported valuable in treating and detecting tuberculosis, malaria, and other illnesses, like the one associated with the Zika virus.

Related to mosquito transmission diseases, the Zika virus (ZIKV) has raised international public health concerns due to its associated symptoms and syndromes. Furthermore, ZIKV diagnostic methods face real challenges as conventional laboratory diagnostics can only record reliable data in the first seven days of the onset of symptoms.⁷⁵ After that time, the only way to obtain

As mentioned before, target 3.3 aims to eradicate waterborne diseases, which are caused by viruses and bacteria that are ingested through contaminated water, food, or by coming in contact with feces. Two of the most common bacteria related to waterborne diseases are *Escherichia coli* (*E. coli*) and *Salmonella typhi* (*S. typhi*). It has been detected that these two waterborne diseases can be prevented by green synthesized BMNPs.^{75,85–88,125} These bimetallic nanosystems will be detailed in Section 6 where SDG 12 is discussed due to their closer relation to this goal.

According to van Vliet *et al.*, water scarcity refers to the lack of water in quantity and the deficit of water quality.¹²⁶ Their study shows that currently, the world's population suffers from severe



water scarcity from an annual average of 40%, taking into account both water quantity and quality.¹²⁶ Aware of the worldwide population's strong demand for clean water, the UN defined SDG 6 as water and sanitation.¹²⁷ SDG 6 seeks to ensure safe drinking water and sanitation for all by implementing sustainable management of water resources, wastewater, and ecosystems and acknowledging the importance of an enabling environment.¹²⁶ Fig. 4 represents the schematic view of some of the applications for traditionally and green synthesized BMNPs related to this SDG, which will be discussed in this section as a possible solution to improve water quality to achieve SDG 6.

Radioactive cesium (Cs) and chromium (Cr) are undesirable metals found in wastewater industrial systems.^{128,129} Given this and looking for a solution toward the release of Cs derived from the accident at the Fukushima Daiichi Nuclear Power Plant, Shubair *et al.*⁶⁶ synthesized Fe NPs and Cu@Fe BMNPs for Cs removal from aqueous solutions. Sorption batch studies showed that the Cu@Fe BMNP sorption process was kinetically faster than that of monometallic Fe NPs. Furthermore, at low concentrations, Cu@Fe NPs presented higher values of maximum removal of Cs, which can be attributed to Cu coating. The presence of Cu increases the surface area and decreases the aggregation and agglomeration of Fe NPs as it also prevents Fe from oxidation by O₂, increasing their reactivity. In a matrix effect evaluation, it was observed that cation ions could block the electron transfer from nanoparticle cores, affecting Cs removal. Nevertheless, it was also observed that Cu@Fe BMNPs selectively adsorbed Cs in the presence of competing cations (Na⁺, K⁺, Mg₂⁺, and Ca₂⁺), indicating that Cu@Fe NPs could be used as efficient materials for Cs removal from contaminated water.¹²⁹ Due to its high toxicity and related exposure to health

risks, the EPA has defined 0.1 mg L^{-1} as the maximum acceptable amount of Cr in drinking water.¹³⁰ Due to this, Das *et al.*³⁰ investigated the catalytic properties of AuAg NPs decorated on L-cysteine functionalized reduced graphene oxide (L-Cys-rGO) sheets toward the peroxidase mimic oxidation of 3,3',5,5'-tetramethylbenzidine (TMB) in the presence of hydrogen peroxide (H_2O_2) as an easy detection of Cr(VI) in an aqueous medium. The detection by the colorimetric nanostructured probe was possible as Cr(VI) ions promoted the decomposition of H_2O_2 to $\cdot\text{OH}$ radicals in an acidic medium which enhanced the oxidation of colorless TMB to its blue colored oxidation product (oxTMB). At the end of the evaluation, it was concluded that the colorimetric sensor exhibited excellent sensitivity and selectivity toward detecting Cr(VI) ions with an LOD value of $26.39 \text{ }\mu\text{M}$. This LOD value was lower than the ones obtained by monometallic nanocomposites Au/L-Cys-rGO and Ag/L-Cys-rGO.

Apart from Cr and Cs, metals like lead (Pb), zinc (Zn), copper (Cu), and manganese (Mn) are found as pollutants in water bodies. Adewoye *et al.*⁷⁷ conducted a study where the aforementioned heavy metals were adsorbed by green synthesized AuAg NPs. Synthesis was mediated by *E. crassipies* plants, using silver nitrate and gold chloride as precursor salts. Samples containing metals were obtained directly from pharmaceutical effluent collected from a Nigerian point of discharge to confirm the adsorption properties and capacity of the synthesized BMNPs. The obtained nanostructured particles were found to be both spherical and cubic-shaped. For the study, different amounts of adsorbent material powders were loaded in the effluent sample (1 mg, 5 mg, and 10 mg). The adsorption tests were carried out using an Inductive Coupled Plasma Atomic Emission Spectroscopy (ICP-OES), quantifying the metal concentration of Pb, Zn, Cu, and Mn before and after the mixing of the effluent and the adsorbent material; mixing was performed for 24, 48, and 72 hours. In general, the adsorption of metallic ions was increased with the contact time for the three studied metals; however, a direct increase of adsorption with respect to adsorbent material concentration in the same amount of time was only found in Cu removal. For Zn removal, 61.13%, 34.39%, and 58.12% ionic concentrations were achieved using 1, 5, and 10 mg of AuAgNPs, respectively. The highest removal efficiency was achieved for Pb, adsorbing 93.37% of its ions with a contact time of 72 h between the sample and 1 mg of AuAg NPs. In the case of manganese, with a contact time of 48 h with the NPs, removal percentages were 51.7%, 19.60%, and 38.05% for 1, 5, and 10 mg of adsorbent material, respectively. The study showed that the size and morphology of bimetallic nanoparticles as adsorbent materials are directly linked to their rate and removal capacity of metallic wastes in water. Apart from this, the recuperation of these nanostructured materials should be further studied to make them feasible in real-life applications.

5.2 Pharmaceutical waste degradation

Moving on to the degradation of chemical compounds in water, it has been detected that antibiotics from different sources have

been released into nature, leaving an important environmental footprint.^{131–133} According to Dong *et al.*,⁶⁷ one of the most commonly detected antibiotics in the environment is tetracycline (TC). They reported the synthesis of FeNi NPs to remove TC under various water conditions. Their results showed that FeNi NPs were efficient materials for TC removal, following adsorption and degradation mechanisms. Furthermore, it was confirmed that these mechanisms were associated with the active sites of FeNi NPs and that the stability of the NPs in dispersion was due to electrostatic repulsion providing many available sites, resulting in a fast decrease of TC at the beginning of the degradation process for relatively low concentrations of TC, 50 and 100 mg L^{-1} .

In this view, Ravikumar *et al.*⁷⁸ synthesized FeNi NPs by a green synthesis method and reported similar outcomes to Dong *et al.*⁶⁷ Both FeNi bimetallic systems showed a spherical shape, a size between 100 and 130 nm and removed TC by absorption and degradation mechanisms. Ravikumar *et al.* showed that a *Pomegranate* extract could substitute NaBH_4 as a reducing agent for this nanosystem. Another advantage was that the green synthesized bimetallic system was coated with carboxymethyl cellulose (CMC), which led to a better dispersion at higher pH values compared to the traditionally synthesized FeNi bimetallic system.^{67,78} Moreover, Ravikumar *et al.* evaluated TC removal in three different matrices and found that the lake water matrix presented the lowest percentage of TC reduction, followed by ground and distilled deionized water matrices. Additionally, they studied the residual toxicity of TC by-products after interacting with FeNi NPs. Viability results showed that TC byproducts had less environmental toxicity than TC without BMNP interaction.⁷⁸ A similar approach was reported by Gopal *et al.*, who also used *Pomegranate* peel to synthesize FeCu ⁷⁹ and Fe@Pd ⁸⁰ bimetallic systems and their respective composites using bentonite (Bt) as supporting materials (Bt-FeCu NPs and Bt-Fe@Pd NPs) for antibiotic TC remediation. In both cases, bentonite composites presented a higher degree of TC reduction than pure bimetallic systems. This behavior is due to bentonite acting as a supporting material and effectively decreasing the aggregation of BMNPs and, consequently, keeping a relatively high surface area. Furthermore, both composites possessed higher stability than BMNPs for prolonged times. In view of these results, the evaluation of pH, residual toxicity, and matrix effects was performed for Bt-FeCu NPs and Bt-Fe@Pd NPs. Experimental results showed the two composites presented approximately the same range of TC effective degradation at both acidic and neutral pH. Nevertheless, a large amount of Bt-Fe@Pd NPs was necessary to achieve this activity compared to Bt-FeCu NPs. Moreover, the outcomes of residual toxicity and removal studies suggested the environmental safety and efficacy of Bt-FeCu NPs and Bt-Fe@Pd NPs for antibiotic removal. Finally, from TC removal studies on different natural water systems, composite Bt-FeCu NPs showed a greater removal in lake water and Bt-Fe@Pd NPs in groundwater. This indicates their potential use towards TC environmental pollution.



5.3 Pesticide removal from water

As mentioned previously, pesticide pollution is considered a global health problem. The spread of pesticides in soil and water systems concerns the scientific community and international organizations. In this regard, the EPA implemented regulations to protect the consumption of drinking water with pesticide contamination.¹³⁴ In view of this, Rosbero and Camacho⁸¹ synthesized star-like Ag/Cu NPs by a co-reduction method using aqueous leaf extracts of *Carica papaya* for water purification applications to degrade chlorpyrifos pesticide. The results of pesticide degradation showed that the star-like Ag/Cu NPs converted chlorpyrifos to less toxic and non-mutagenic degradation products, such as 3,5,6-trichloropyridinol (TCP) and diethylthiophosphate (DETP). Conclusively, using G-BMNPs for chlorpyrifos degradation could be considered a sustainable environmental remediation since it can be applied at room temperature without the need for UV light activation.⁸¹ Furthermore, arsenic (As) is one of the compounds present in pesticides. Lin *et al.*⁸² synthesized calcined Fe/Pd NPs for As(III) removal from wastewater. Calcination caused an increase in the specific surface area, which provided additional active adsorption sites for both As(III) and As(V). Furthermore, it was observed that an alkaline pH and a higher adsorbent dose improved the removal efficiency of As(III). In this study, calcined-Fe/Pd NPs provided a complete removal (100%), proving to be a suitable composite for As removal from wastewater.

Some previously described studies embark on water and sanitation and contribute to achieving SDG 12: responsible consumption and production.^{128,131} This is important as the current lifestyles do not involve responsible consumption and production patterns. Due to the relevance of this sustainable goal, specific examples of BMNPs related to SDG 12 are going to be discussed in the following section.

6 Case studies of bimetallic nanoparticles in relation to SDG 12: responsible consumption and production

It has been detected that one of the main causes of food loss is microbiological food spoilage, which impacts food quality and shelf life. Food spoilage results from microbiological, chemical, or physical changes in food products, making them unsuitable for consumption.¹³⁵ These issues may result in a variety of infections and/or intoxications for consumers.¹³⁶ Additionally, the UN reported that each year, approximately 1.3 billion tons of all food produced worldwide is lost or wasted. Concerned by these undesirable consumption and production patterns, SDG 12.3 stated the goal to halve per capita global food waste at the retail and consumer levels and reduce food losses along production and supply chains, including post-harvest losses by 2030.¹³⁷ In the face of this problem, BMNPs synthesized by traditional and green synthesis methods could be taken as a strategy to increase the shelf life, avoiding food loss. Some of

these solutions are shown in Fig. 5 and are discussed in the following section.

6.1 Packaging solutions

It is estimated that around 20–60% of fresh food production suffers losses at various operational levels in the supply chains.¹³⁸ Attending this, recent studies have focused on designing and creating food packaging films to avoid losses in the production chain. A case in point is the work of Arfat *et al.*, who synthesized three different nanocomposite films using agar,⁶⁸ polylactide (PLA)⁶⁹ and linear low-density polyethylene (LLDPE)⁷⁰ as polymeric matrices and loaded them with Ag/Cu NPs (Ag/Cu NPs-agar, Ag/Cu NPs-PLA, and Ag/Cu NPs-LLDPE, respectively). In the case of Ag/Cu NPs-PLA and Ag/Cu NPs-LLDPE composites, cinnamon essential oil was loaded together with BMNPs. The results showed that adding NPs caused a change in film opacity, impeding the transmission of UV and visible light through the films. These changes in the color parameters were attributed to the surface plasmon resonance of the Ag/Cu NPs, making the composite films feasible as packaging for easily-oxidizing food due to light radiation. Furthermore, the three composites improved the oxygen barrier properties and antimicrobial activity, which could be explained by the release of Ag⁺ and Cu²⁺ ions into the bacterial membrane and their interaction with enzymes and organelles, producing cellular death. To corroborate the antibacterial activity in real samples, microbial validation tests were performed in chicken samples for Ag/Cu NPs-PLA and Ag/Cu NPs-LLDPE composites. Both exhibited remarkable growth reduction against *Listeria monocytogenes* (*L. monocytogenes*), *Salmonella Typhimurium* (*S. Typhimurium*), and *Campylobacter jejuni* (*C. jejuni*). From these three, the nanocomposites showed the greatest bacterial

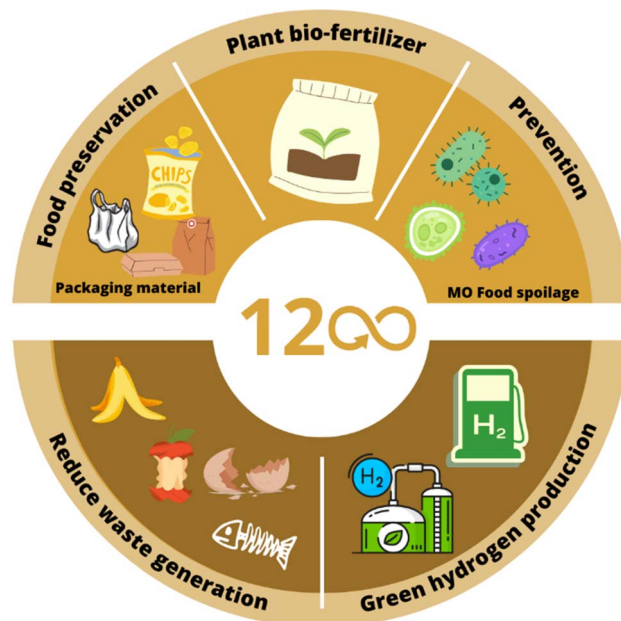


Fig. 5 Applications of traditional and green synthesized BMNPs related to SDG 12: responsible consumption and production.



Gulbagca *et al.*⁹¹ fabricated Pd-Ag NPs, with spherical morphology and a particle size around 7 nm, by a green route using a *Nigella Sativa* plant extract. The effects of Pd-Ag NP concentration, temperature, NaBH₄ concentration, and pH of the system were tested towards hydrogen obtention. At a constant NaBH₄ concentration (125 mM), the concentrations of Pd-Ag NPs were 2.25, 4.50, 6.75, and 9 mM, and the process was done for 30 min. Even at the lowest BMNP concentration, the system released around 150% more volume of H₂ than the blank test, *i.e.*, without BMNPs, showing high catalytic activity and better performance compared to the work of Darabi,⁹⁰ producing 10 mL more than the mentioned work. On the other hand, while having a constant Pd-Ag NP concentration and varying NaBH₄ concentrations, the obtained volume of H₂ reached around 30 mL in 30 min, which is around three times more volume than the blank test at the same time. A temperature test showed that the amount of obtained H₂ increased with respect to the system's temperature. This test was also useful to determine the E_a , change in Gibbs energy (ΔG), and change in entropy (ΔS) of the system, which were 24.51, 27.01, and $-183.15 \text{ kJ mol}^{-1}$, respectively. In the case of pH studies, it was found that at lower pH values, the yield of H₂ obtention was favored, with an optimum pH value of 2.82. In

In another strategy, Mallikarjuna and team⁹⁴ synthesized Pd–Ag BMNPs that were combined with rGO, resulting in Pd–Ag/rGO heterostructures. As in the previous examples, the synthesis of the BMNPs was carried out following a green synthesis route, using a stevia leaf extract. TEM images revealed that the spherical Pd–Ag NPs were distributed on the rGO sheets, with particle sizes ranging from 5–15 nm. In this study, water was used instead of NaBH₄ as the hydrogen source, together with methanol as a scavenger agent. Also, the study tested the photocatalytic activity of the synthesized nanostructures using an artificial solar light simulator. The rGO, Pd/rGO, and Pd–Ag/rGO systems were tested, where the sole rGO system produced the lowest yield of H₂ of 620 $\mu\text{mol g}^{-1}$. The presence of Pd/rGO increased this yield to around 2400 $\mu\text{mol g}^{-1}$, while the system with the highest H₂ production yield was provided by the Pd–Ag/rGO heterostructures with around 5800 $\mu\text{mol g}^{-1}$ of produced H₂ due to a relatively higher surface area, lower band gap and lower electron–hole recombination rate.⁹⁴ The yield of H₂ evolution was found to be non-linear between doses of 2.5–10 mg Pd–Ag/rGO since it reached a maximum value of 5802 $\mu\text{mol g}^{-1}$ of H₂ product with 5 mg of Pd–Ag/rGO, which decreased by increasing the mass of Pd–Ag/rGO. The addition of methanol as a scavenger agent was also tested, and it was found that the optimum concentration was around 5% v/v, since, beyond this value, the production of hydrogen decreases because of generated methanol byproducts. Reusability tests were also carried out, where the Pd–Ag/rGO photocatalytic system decreased its performance only slightly after five cycles of use.

These coinage metals led to an increase of intensity in the SERS signal with respect to the sole Cu system, together with the enhanced stability for several weeks of the system due to the presence of Au. Stability obstacles may also be overcome by modifying either their synthesis or implementation method. An example is the incorporation of metallic NPs on solid substrates, such as silica, alumina, or carbon.¹³ Zhuang and colleagues¹⁶⁴ prepared a hydrogen obtention solution based on a carbon fiber paper with Pd-Au NPs loaded into it, using the *Eucalyptus* plant as a reducing and stabilizing agent. Long-term potential cycles and potentiostatic electrolysis were used to confirm the stability and durability of the system due to the firm anchoring of the NPs to the substrate. Stability may also be achieved by incorporating a stabilizing agent in the synthesis method,¹³ such as PVP,⁷⁴ CMC,⁷⁸ and chitosan.⁹⁶ Although plant-synthesized BMNPs face reproducibility challenges and their phytochemical nature and properties are normally not well understood, the reducing and stabilizing agents found in the plant extracts may show enhanced stability and low toxicity characteristics.¹⁶⁵ For instance, Sundarrajan *et al.*¹⁶⁶ studied how the *Artocarpus heterophyllus* fruit extract could be used both as a reducing and a stabilizing agent in synthesizing Au@Ag NPs, and their formation mechanism was analyzed. In this case, the mechanism involved an activation, growth, and termination process.¹⁶⁶ The first consisted of the reduction of metal ions by an electronation process, followed by an association of metallic atoms, leading to thermodynamic stability, due to the extract's bioactive components of the extract.¹⁶⁶ Therefore, using stabilizing agents, either from plant extracts or other agents, and comprehending the systems' mechanisms may lead to more stable NP products.

7.4 Challenges of the implementation of BMNPs in real-life sustainability affairs

Apart from scale-up production challenges, real-life conditions play a major role in the effectiveness of nanomaterial-based solutions for SDGs 3, 6, and 12. As seen before, most of the issues related to SDG 3 are from soil remediation techniques. Although the degradation of chemicals such as TCE, chlorophenols, and nitroaromatic compounds has been enhanced by BMNPs, the mechanisms of these nanostructured materials may vary depending on the region and climatic conditions.¹⁶⁷ The latest is due to the presence of organic and metallic compounds, as well as conditions associated with local pH and mean temperatures of the environments to be remediated.¹⁶⁷ Furthermore, nanomaterials may show adverse effects on organisms found in soil, which may lead to negative implications in the seeking of remediation processes, aggravating soil conditions and affecting natural processes.¹⁶⁸ Nanoparticles used in water remediation processes (SDG 6) have been found to be effective in detecting and removing pollutants. Nevertheless, as with soil, natural conditions may affect the processes involved in water remediation. Metallic nanoparticles, specifically the ones with magnetic properties, are found to be unstable under acidic or alkaline conditions, which leads to the loss of their magnetic force and a reduced time of life.¹⁶⁸ This

7.3 Overcoming stability obstacles

As it is approached in this review, one route that can be followed is the bimetalization of the systems, which is a key factor for improving the stability of metallic nanosystems.¹⁶² As an example, metals like Cu may be susceptible to oxidation when exposed to air;¹⁶³ hence, Jiang and colleagues developed Cu–Au nanotubes for their application in SERS detection.¹⁶³

With respect to the measurement of the environmental impact of the application of BMNPs, a study reported by Ulucan-Altuntas *et al.*¹⁷⁴ shows the Life Cycle Assessment (LCA) of the production of FeCu *via* traditional methods, *i.e.*, by using NaBH₄ as a reducing agent. LCA shows that around 87% of the synthesis environmental damage for producing FeCu BMNPs comes from using NaBH₄ in the process.¹⁷⁴ The authors of this report stated that “to remove the borohydride reagent from the method, greener synthesis should be used”, since around 1.87 m³ of wastewater with borohydride concentrations were produced by synthesizing 1 kg of BMNPs.¹⁷⁴ Similar results were obtained in the study performed by Visentin *et al.*,¹⁷⁵ where different production processes of metallic NPs were analyzed

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