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Review Article

The Uses of Various Nanoparticles in Organic Synthesis: A Review

Khaturia S[1], Chahar M[2*], Sachdeva H[3], Sangeeta[1], Mahto CB[2]

[1]Department of Chemistry, School of Sciences, Mody University of Science and Technology, Laxmangarh (Sikar), Rajasthan, India; [2]Department of Chemistry, Nalanda College of Engineering, Chandi, Nalanda, Bihar, India; [3]Department of Chemistry, University of Rajasthan, Jaipur, India

ABSTRACT

Nanoparticles have been widely applied in different areas including, medicine, sensor and catalysis. In our study we have concentrated our work towards the application of the metal nanoparticles in the field of catalysis. Several reports has been found on wide range of application of various supported metal nanoparticles in catalysis including Au, Ag, Pt, Cu, Cd, Ni etc. metals in the form of reduced metals and in compounds forms as heterogeneous catalysis. Nanoparticles have potential for improving the efficiency, selectivity and yield of catalytic processes. Higher selectivity of the nanoparticles towards reaction proceeds through less waste and fewer impurities which could lead to safer technique and reduced environmental impact. In this review we have focused on the developments in new types of green nanocatalysts as well as developments in green catalytic reactions.

Keywords: Nanoparticles; Nanocatalysts; Organic synthesis; Green Nanocatalysts; Green reactions; Nanotechnology.

INTRODUCTION

The last decade has witnessed enormous development in the field of nanoscience and nanotechnology. Several reports show the amazing level of the performance of nanoparticles as catalysts in terms of selectivity, reactivity and improved yields of products. In addition, the high surface-to-volume ratio of nanoparticles provides a larger number of active sites per unit area, in comparison with their heterogeneous counter sites [1,2]. In this review, we focus on green nanocatalysts as well as industrially important green reactions. This article has two parts. The first part involves green nanocatalysts and the second part involves green reactions.

SYNTHESIS OF VARIOUS NANOPARTICLES

Various nanoparticles are shown in Figure 1 and Schemes 1-53 in Tables 1-15.

Calcium oxide nanoparticles

Among various nanoparticles, calcium oxide nanoparticles have

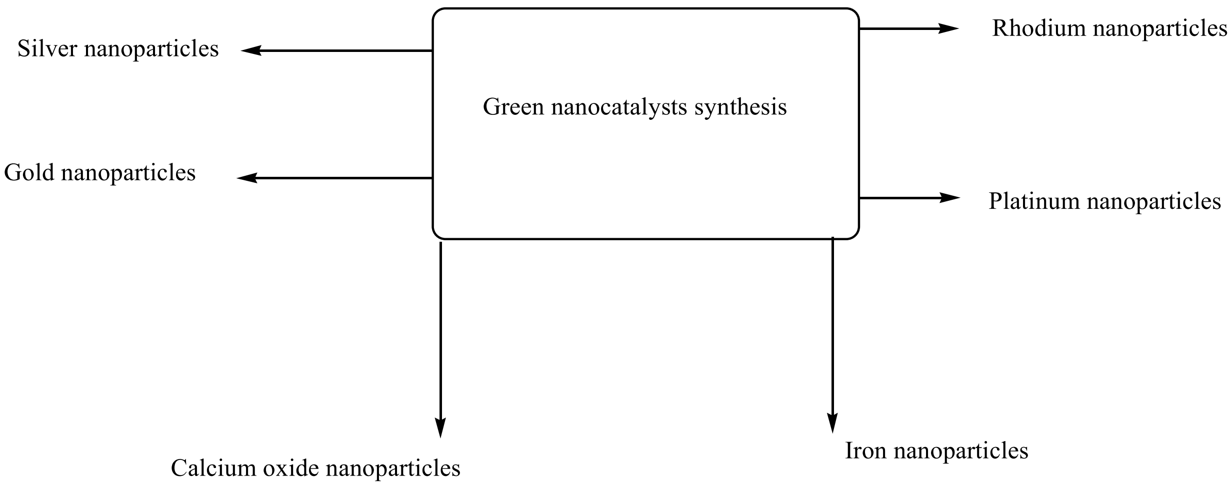

Figure 1: Various Green Nanocatalysts.

*Correspondence to: Mamta Chahar, Department of Chemistry, Nalanda College of Engineering, Chandi, Nalanda, Bihar, India, Tel: 7737075718; E-mail: mamtachahariitd@gmail.com

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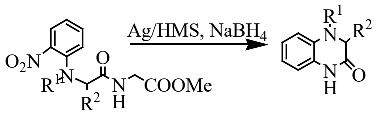
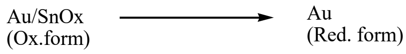
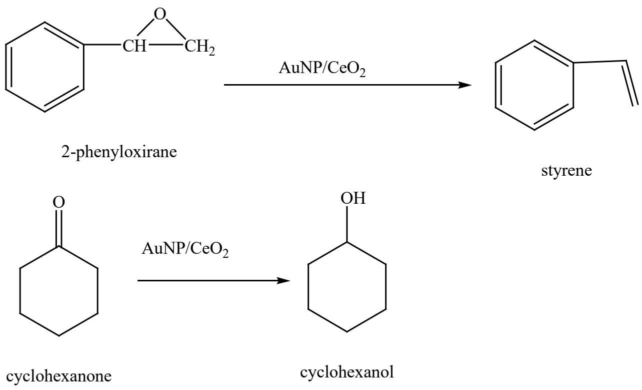
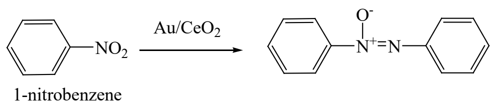
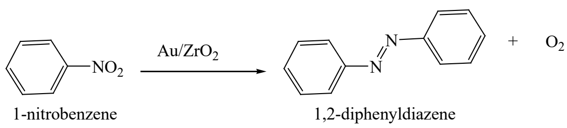
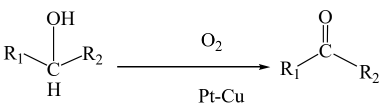
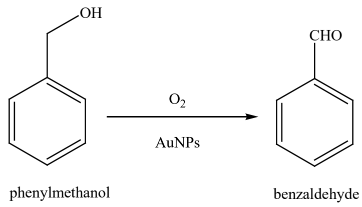
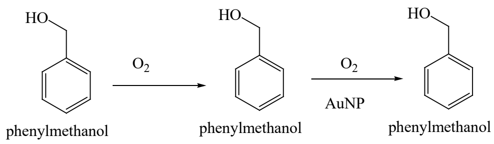
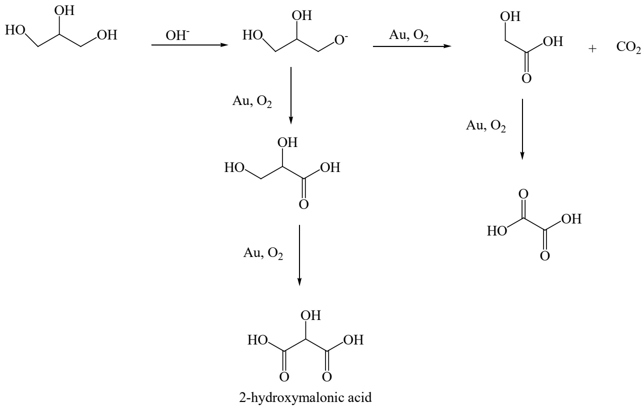
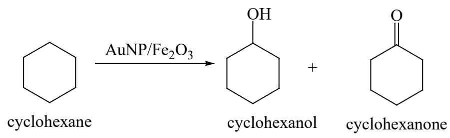
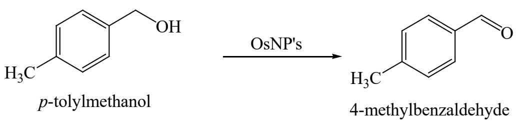
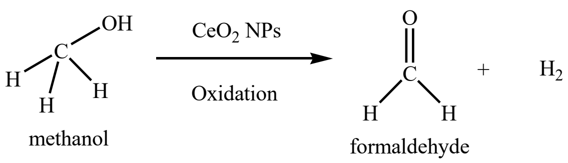
Table 1: Reduction Reactions.

S. No.	Reference No.	Reaction	Nano particles
	85	 **SCHEME 1: Synthesis of Amine-Substituted MCR Scaffolds and Dihydroquinoxalinone Derivatives**	Ag
	86	 **SCHEME 2: Reduction of Methylene Blue dye**	Ag
	87	 **SCHEME 3: Oxygen Reduction Reaction at Gold Nanoparticles**	
	88	 **SCHEME 4: (i) Synthesis of styrene from 2-phenyl oxirane (ii) Synthesis of cyclohexanol from cyclohexanone**	Au
	89	 **SCHEME 5: Selective reduction of nitrobenzene**	
	90	 **SCHEME 6: Synthesis of 1,2-diphenyldiazine from nitrobenzene**	
	91	 **SCHEME 7:Synthesis of substituted ketone by substituted alcohol**	Pt-Cu alloy

received considerable attention because of their unusual properties and potential applications in diverse fields [3]. Calcium oxide (CaO) itself as cost effective, highly basic, non-corrosive, environment friendly, and economically benign, that can be regenerate and reused. Also, they require only mild reaction conditions to produce high yields of products in short reaction times, in comparison with traditional catalysts [4-6]. Many researchers reported that calcium oxide nanoparticles as an active catalyst in many chemical transformations such as adsorption of Cr (VI) from aqueous solutions [7], biodiesel trans-esterification [8-19], removal of toxic heavy metal ions in water [20] and artificial photosynthesis [21] and the degradation of bromocresol green [22], purification of vehicle gas exhaust [23]. In accordance with the above mentioned consequence of nanoparticles in catalysis, and the significance of

Table 2: Oxidation reactions.

S. No.	Reference No.	Reaction	Nano particles
1.	92	 **SCHEME 8: Synthesis of benzaldehyde by phenyl methanol**	Au
2.	93	 **SCHEME 9: Selective oxidation of benzyl alcohol**	
3.	94	 **SCHEME 10: Synthesis of 2-hydroxymalonic acid**	
4.	95	 **SCHEME 11: Synthesis of cyclohexanol and cyclohexanone from cyclohexane**	
5.	96	 **SCHEME 12: Synthesis of sustituted benzaldehyde by substituted alkanol**	**Os& Ir**
6.	97	 **SCHEME 13: Synthesis of Formaldehyde by methanol**	CeO_2

highly substituted pyridines as privileged medicinal scaffolds.[24] Calcium oxide nanoparticles, as an efficient, non-explosive, eco-friendly, non-volatile, recyclable and easy to handle catalyst, can be used in the catalysis of many organic transformations.

Preparation of CaO nanoparticles

NaOH (1 g) was added to a mixture of ethylene glycol (12 ml) and $Ca(NO_3)_2$. $4H_2O$ (6 g) and the solution stirred vigorously at room temperature for 10 min; the gel solution was kept about 5 h at

Table 3: Conversion of organosilanes to silanols.

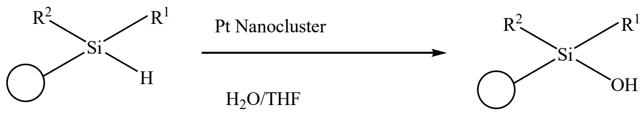
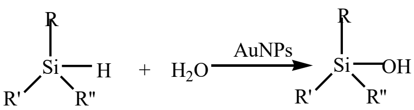
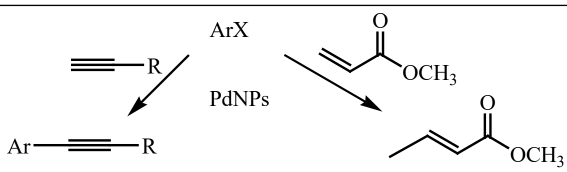
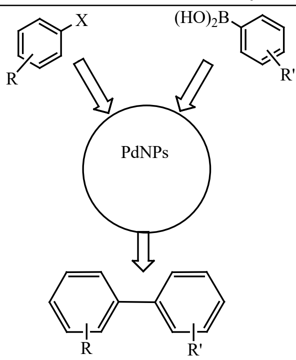
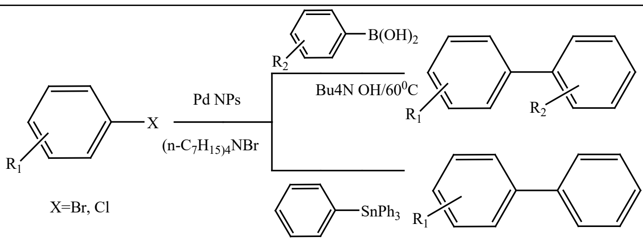
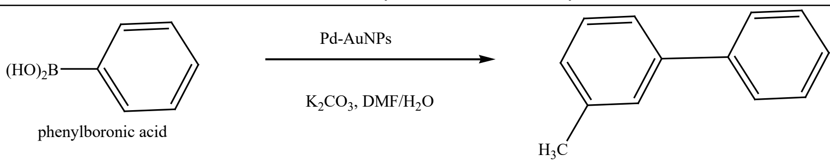
S. No.	Reference No.	Reaction	Nano particles
	98	 **SCHEME 14: Synthesis of Silanols from organosilanes**	Pt
2	99	 **SCHEME 15: Synthesis of Silanols**	Pd
3	100	 **SCHEME 16: Synthesis of Silanols**	Au

Table 4: Suzuki cross-coupling Reactions and Sonagashira Reaction.

S. No.	Reference No.	Reaction	Nano particles
	101	 **SCHEME 17:Synthesis of methyl cinnamate**	Pd
	102, 103, 104, 105	 **SCHEME 18: Synthesis of biaryls**	
	106	 **SCHEME 19: Synthesis of substituted biaryls**	
•	107	 **SCHEME 20: Synthesis of substituted biaryls by Phenyl boronic acid**	Au–Pd alloy

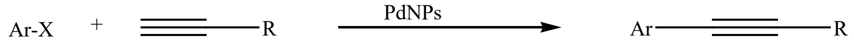
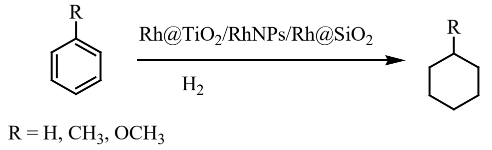
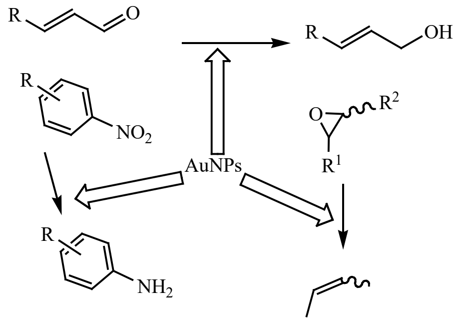
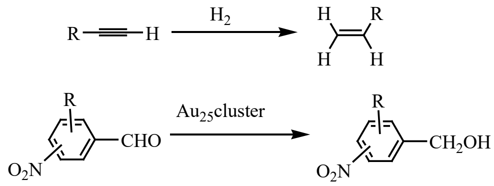
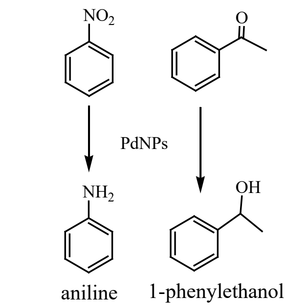
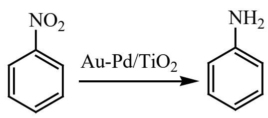
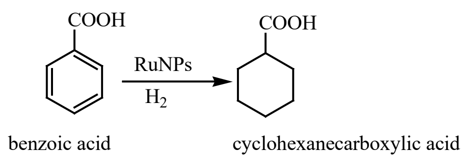
•	108	 **SCHEME 21: Synthesis of substituted Alkyne**	Pd

Table 5: Hydrogenations.

S. No.	Reference No.	Reaction	Nano particles
1.	109, 110	 **SCHEME 22: Synthesis of substituted cyclohexanes**	Rh
2.	111	 **SCHEME 23: Synthesis of substituted anilines**	Au
3.	112	 **SCHEME 24 Synthesis of substituted alkenes**	Au nano cluster
4.	113	 **SCHEME 25:Synthesis of Aniline and 1-phenylethanol**	Pd
5.	114	 **SCHEME 26: Synthesis of Aniline**	Au-Pd
	115	 **SCHEME 27: Synthesis of cyclohexanecarboxylic acid**	Ru

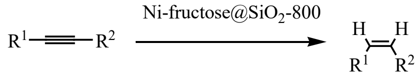
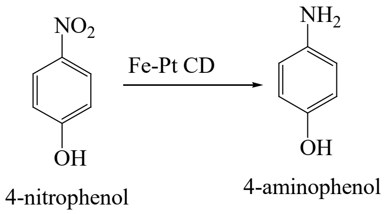
116	 SCHEME 28: Synthesis of Alkene derivatives	Ni
117	1-Hexene —Pd-Ru NPs→ n-Hexane + cis-2-Hexene + trans-2-Hexene **SCHEME 29:Synthesis of hexanes**	RuPd
118	 **SCHEME 29: Synthesis of 4-aminophenol**	FePt

Table 6: Ullmann Reaction.

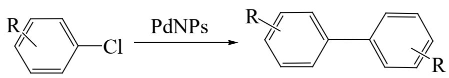
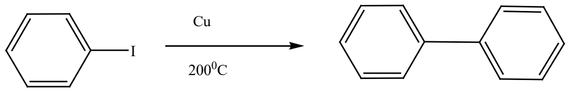
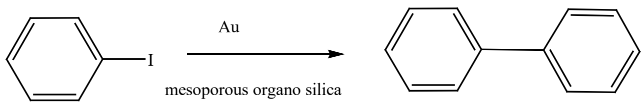
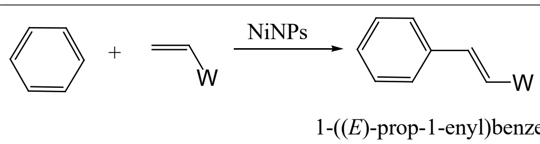
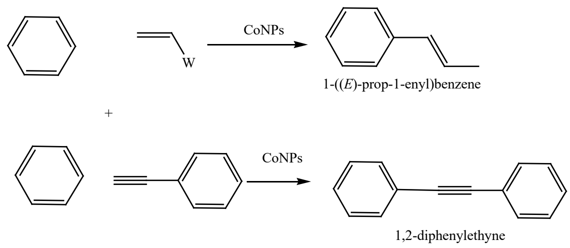
S. No.	Reference No.	Reaction	Nano particles
	119, 120, 121	 **SCHEME 30:Synthesis of biaryl derivatives**	Pd
	122	 **SCHEME 31: Synthesis of biaryls**	Cu
	123	 **SCHEME 32: Synthesis of biaryls**	Au

Table 7: Heck cross-coupling Reaction.

S. No.	Reference No.	Reaction	Nano particles
124		 **SCHEME 33: Synthesis of 1-((*E*)-prop-1-enyl)benzene**	Ni
125		 **SCHEME 34:Synthesis of 1-((*E*)-prop-1-enyl)benzene and 1,2-diphenylethyne**	Co

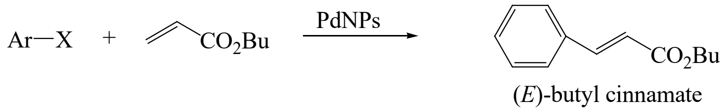
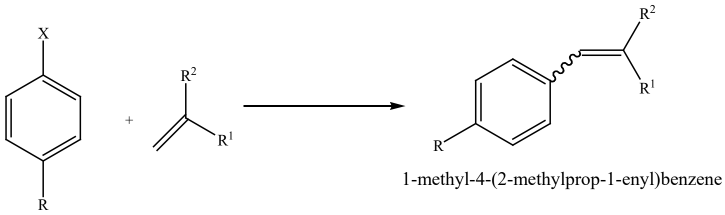
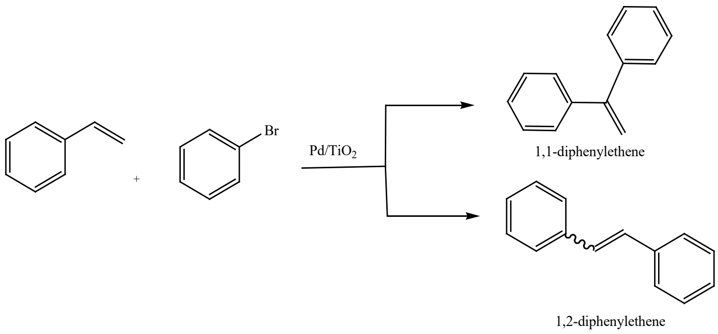
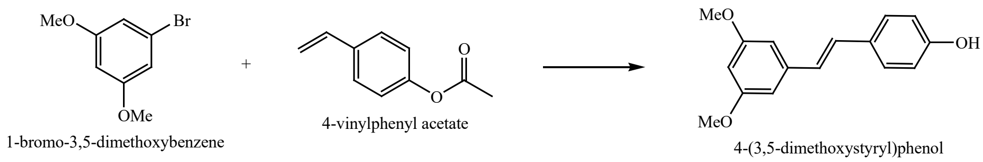
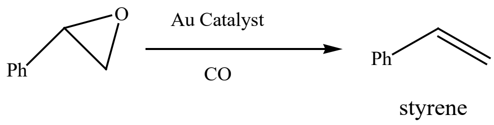
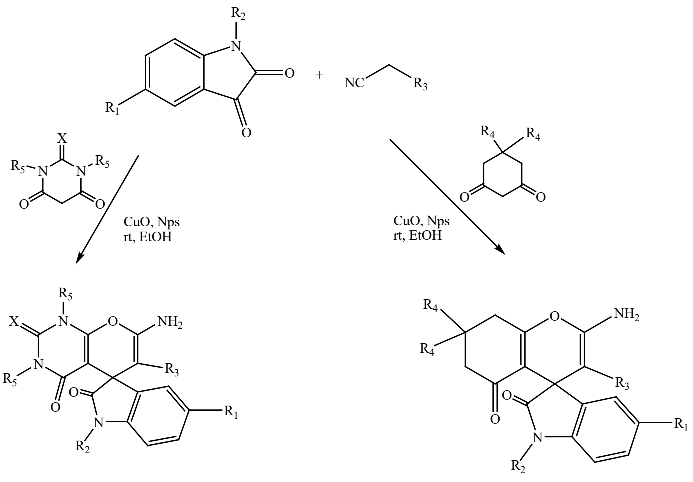
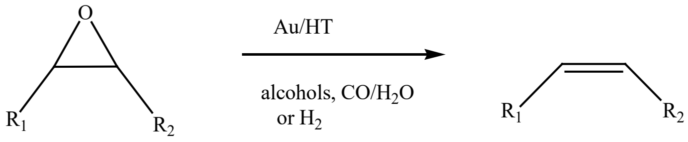
•	126	 **SCHEME 35 : Synthesis of butyl cinnamate**	Pd
	127	 **SCHEME 36: 1-methyl-4-(2-methylprop-1-enyl)benzene**	
	128, 129	 **SCHEME 37: Synthesis of 1,1-diphenyl etane and 1,2-diphenylethane**	
	130	 **SCHEME 38:Synthesis of 4-(3,5-dimethoxystyryl)phenol**	

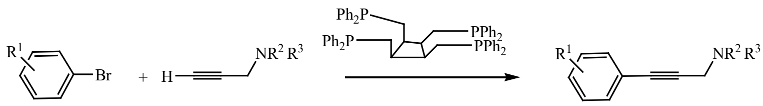
Table 8: Deoxygenation Reaction.

S. No.	Reference No.	Reaction	Nanoparticles
	131	 **SCHEME 39: Synthesis of Styrene**	Au
	132	 **SCHEME 40: Synthesis of Spirooxindoles**	CuO

	Reference No.	Reaction	Nano particles
	133		Au

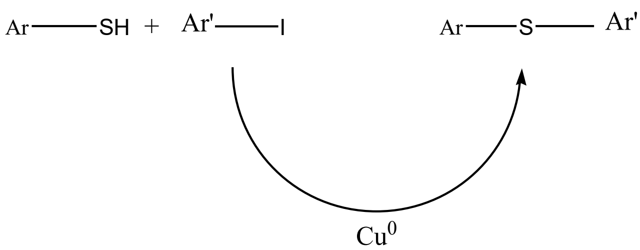
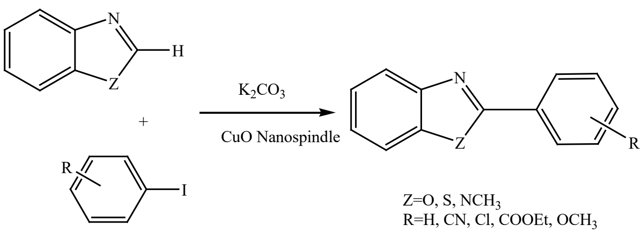
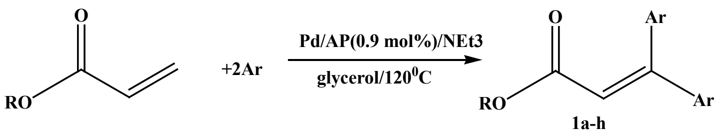
SCHEME 40 : Synthesis of substituted but-2-ene

Table 9: Alkynylation of Aryl Halides.

S. No.	Reference No.	Reaction	Nano particles
1	134		Pd

Scheme 41: Synthesis of *N,N*-dialkyl-3-*p*-tolylprop-2-yn-1-amine

Table 10: Arylations and Diarylations.

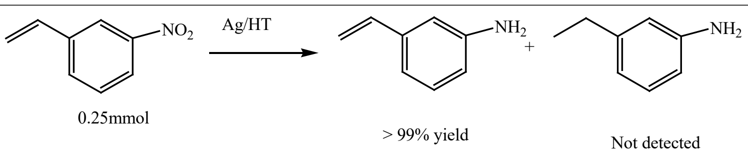
S. No.	Reference No.	Reaction	Nano particles
	135		FeCu
	136		CuO
	137		Pd

SCHEME 42 :Synthesis of diaryl Sulfane

SCHEME 43 : Synthesis of 2-phenylbenzo[d]thiazole, 2-phenylbenzo[*d*]oxazole and 1-methyl-2-phenyl-1*H*-benzo[*d*]imidazolederivatives

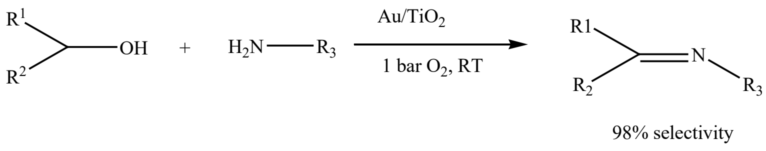
SCHEME 44 : alkyl 3,3-diphenylacrylate

Table 11: Deoxygenation of Epoxides.

S. No.	Reference No.	Reaction	Nanoparticles
1.	138		Au, Ag & Cu

SCHEME 45: Synthesis of 3-vinylbenzenamine and3-ethylbenzenamine

Table 12: Oxidative Coupling of Alcohols.

S. No.	Reference No.	Reaction	Nano Particles
1.	139		Au

SCHEME 46:Synthesis of *N*-(propan-2-ylidene)methanamine

Table 13: Esterification of Alcohols.

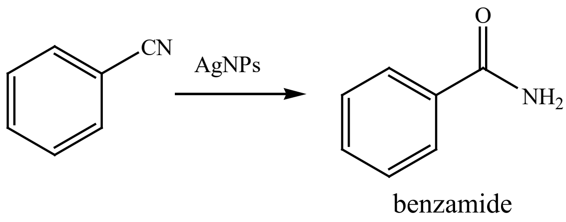
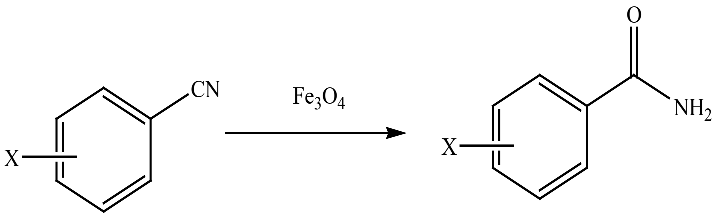
S. No.	Reference No.	Reaction	Nanoparticles
1	140	 **SCHEME 47 : Synthesis of benzamide**	Ag
2	141	 **SCHEME 48: Synthesis of substituted benzamide**	Fe_3O_4

Table 14: Hydration of Nitriles.

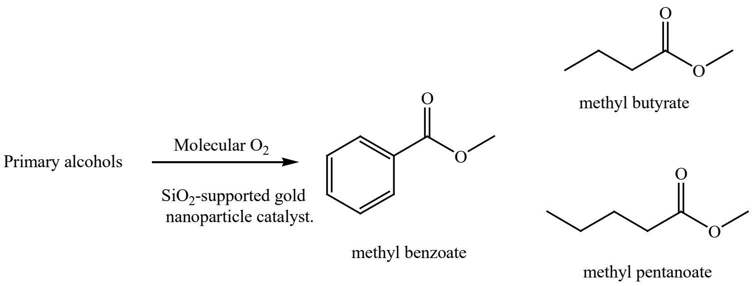
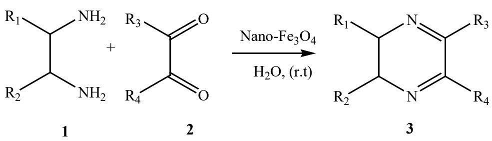
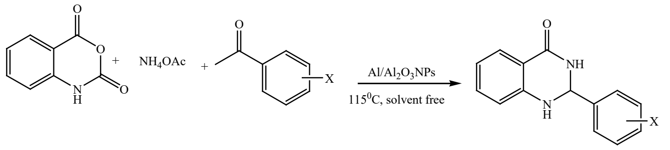
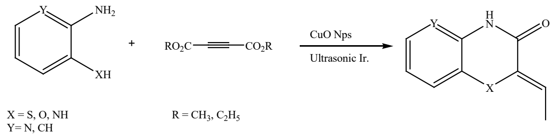
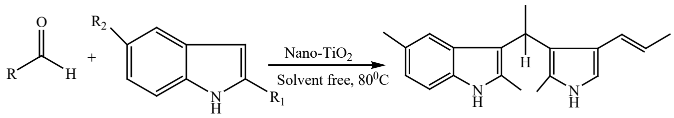
S. No.	Reference No.	Reaction	Nano particles
1	142	 **SCHEME 49: Synthesis of methyl esters**	Au

Table 15: Additional Organic Synthesis Reactions.

S. No.	Reference No.	Reaction	Nano particles
1. 1.	143	 **SCHEME 50 : Synthesis of tetraalkylpyrazine**	Fe_3O_4

2. 2.	80	 SCHEME 51: Synthesis of 2,3-dihydro-2-phenylquinazolin-4(1*H*)-one	Al/Al_2O_3
	144	 SCHEME 52: Synthesis of (Z)-2-ethylidene-2H-pyrido[3,2-b][1,4]oxazin-3(4H)-one and (Z)-2-ethylidene-2H-benzo[b][1,4]thiazin-3(4H)-one	CuO
	145	 SCHEME 53: Synthesis of 2,5-dimethyl-3-(1-(2,5-dimethyl-1*H*-indol-3-yl)ethyl)-1*H*-indole	TiO_2

static state. Afterwards, it was washed using water and dried under vacuum drying. Finally, the prepared CaO nanoparticles were calcinated at 700°C for 3 h [23].

Iron nanoparticles

Iron nanoparticles have been synthesized using polymers as capping agents in water as green solvent as well as other types of green methods. A green synthesis of iron nanoparticles has been prepared by using tea polyphenols without the use of additional polymers and surfactants [24]. The iron nanoparticles are used to catalyze the hydrogen peroxide for treatment of organic contamination. Iron nanoparticles have been used as environmentally benign catalysts for alkene and alkyne hydrogenations [14]. Iron nanoparticles that have been synthesized used as catalysts for environmentally benign alkene and alkyne hydrogenation reactions [25].

Rhodium nanoparticles

CaO and co-workers have reported the catalytic activity of rhodium nanoparticles deposited on modified SiO_2 for hydrogenation of nitrile butadiene rubber (NBR) [26]. Kang et al. investigated the morphology control synthesis of Rh nanostructures for cancer treatment [27]. Rhodium nanoparticles were used in Suzuki-Miyamuri reaction and hydrogenation of Benzene by Gniewek and coworkers [28].

Rh nanoparticles can be synthesized using a variety of green methods such as the hydrogen reduction method in water as solvent, ethanol reduction method in an ethanol water mixture in which the ethanol can be rotovaped, and several other green methods. Rhodium nanoparticles adsorbed onto titanium dioxide supports are synthesized using water as the solvent [29].

Zinc oxide nanoparticles

Zinc oxide nanoparticles (ZnO NPs) are cost effective and relatively less toxicity, significant biocompatibility reveal their remarkable biomedical applications, such as anticancer, drug delivery, antibacterial, diabetes treatment and anti-inflammation [30-35].

Due to the strong UV absorption properties of Zinc Oxide, they are used in cosmetics and sunscreen [36-38]. In addition, these particles also show excellent luminescent properties and used for bioimaging [39,40].

Zinc is the most important component of various enzyme systems, it takes part in body's metabolism and plays essential roles in proteins and nucleic acid synthesis, hematopoiesis, and neurogenesis [41,42].

Johnson et al. developed a new method for the green synthesis of ZnO nanoparticles. In this method, a new leucine-based diamine amphiphile was synthesized and self-assembled. In the presence of Zn^{2+} ions, the leucine-based diamine amphiphile assembled into nanofibers that efficiently formed ZnO nanoparticles on heating with $Zn(CH_3COO)_2$ [43].

Platinum nanoparticles

Platinum nanoparticles (PtNs) possess a wide range of properties that can be used for various applications such as catalysts in organic catalysis, fuel cells, hydrogen storage, electrical conductivity, optics and nonlinear optics, coating, plastics, textile, biosensors and biomedicine [44-50]. Engelbrekt and co-workers demonstrated the synthesis of PtNPs using a variety of green methods such as the hydrogen reduction method in water as solvent, ethanol reduction method in an ethanol- water mixture. Monodisperse green Pt nanoparticles were synthesized by using glucose as the reducing agent and starch as the protective agent [51,52]. This synthesis method is environmentally friendly, highly reproducible, and easy to scale up. These nanocatalysts were tested for reduction and oxidation reactions and were found to have high catalytic activity. Moreover, these Pt nanoparticles are stabilized with ionic liquids and used as catalysts for four-electron reduction of dioxygen to water [53].

Gold nanoparticles

In recent years, AuNPs had attracted an immense interests in different fields of science, due to their unique features such as high X-ray absorption coefficient, ease of synthetic strategy, enabling precise control over the particle's physico-chemical properties, strong binding affinity to thiols, disulfides and amines, unique tunable optical and distinct electronic properties [54-60]. The optical-electronics properties of gold nanoparticles are being explored extensively for high technology applications such as sensory probes, electronic conductors, therapeutic agents, organic photovoltaics, Fuel cells, drug delivery in biological and medical applications, and catalysis [61-65].

Itoh et al. investigated the synthesis and functions of gold nanoparticles with ionic liquids based on the imidazolium cation.

At room temperature green imidazolium-based ionic liquids such as 1-butyl-3-methylimidazolium hexafluorophosphate are used as liquid media for the synthesis of gold nanoparticles which can be used in dyes [66].

The gold nanoparticles were prepared by the addition of $HAuCl_4$ to green tea leaves extract at room temperature. The synthesis of the Au nanoparticles does not involve any toxic chemicals/ organic solvents so it is a green synthetic process. The gold nanoparticles are used as catalysts for the reduction of methylene blue dye [67]. Au nanoparticles have been synthesized by a green photo catalytic method in which the synthesis is conducted in water [68]. Calcium-alginate stabilized gold nanoparticles are prepared using a photochemical green synthetic method [69]. Zhan and co-workers used Au nanoparticles as catalysts for the 4-nitrophenol reduction reaction. They have prepared Gold/TS-1 nanoparticles using two green routes which are sol-immobilization method and adsorption reduction method [70]. This gold nanoparticle catalyst show excellent performance for the propylene oxidation reaction.

Silver nanoparticles

Silver nanoparticles have commercialization applications for instance, sterilizing nanomaterials in consuming and medical products, textiles, food storage bags, refrigerator surfaces, and personal care products [71-74]. Additionally, they show optical, thermal, and catalytic properties and antimicrobial ability [75-79].

Silver nanoparticles have been synthesized using several green methods such as the seed-mediated growth method, in the presence of ionic liquids, and other reduction methods such as hydrazine reduction method, and sodium borohydride reduction method. Ag nanoparticles have been synthesized by a green photocatalytic method in which reaction is conducted in water [68]. Calcium-alginate stabilized silver nanoparticles are prepared using a photochemical green synthetic method [69]. These nanoparticles are used as catalysts for the 4-nitrophenol reduction reaction.

Aluminium nanoparticles

Solvent-free methods as well as methods involving the use of water as solvent have been used to synthesize aluminum oxide nanoparticles. Aluminum oxide nanoparticles are synthesized in water as the solvent which makes it a green nanocatalyst [80].

Bimetallic nanoparticles

Bimetallic nanoparticles (Figure 2) have been prepared by the ethanol reduction method, hydrogen reduction method, and other green methods. These nanoparticles have been used as catalysts in several organic chemistry, including, oxidation of carbon monoxide in aqueous solutions, hydrogenation of alkenes in organic or biphasic solutions and hydrosilylation of olefins in organic solutions [81,82].

Nickel platinum nanoparticles

Nickel encapsulated by Pt (NiPt) has been synthesized using a green colloidal method [83]. Pt NPs are very expensive as electrocatalysts so the remedy for this is to diminish the cost by the synthesis of Ni-Pt bimetallic nanoparticles.

Gold-palladium nanoparticles

Au-Pd nanoparticles are preared in the absence of organic ligands and adsorbed onto TiO_2 supports and is found to be stable in oxidative catalysis conditions [84]. It was investigated that 70% gold, 30% Pd composition of the bimetallic nanoparticles show the highest catalytic activity for the oxidative catalysis.

Application of various nanoparticles in green reactions

Applications of different nanoparticles in green reactions are brief in Figure 3 and summarized in Tables 1-15 [85-145].

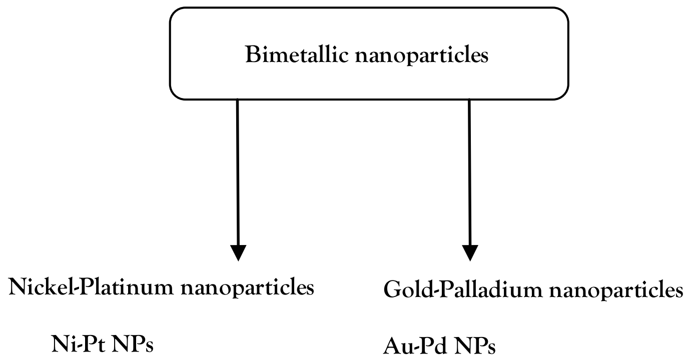


Figure 2: Bimetallic Nanoparticles.

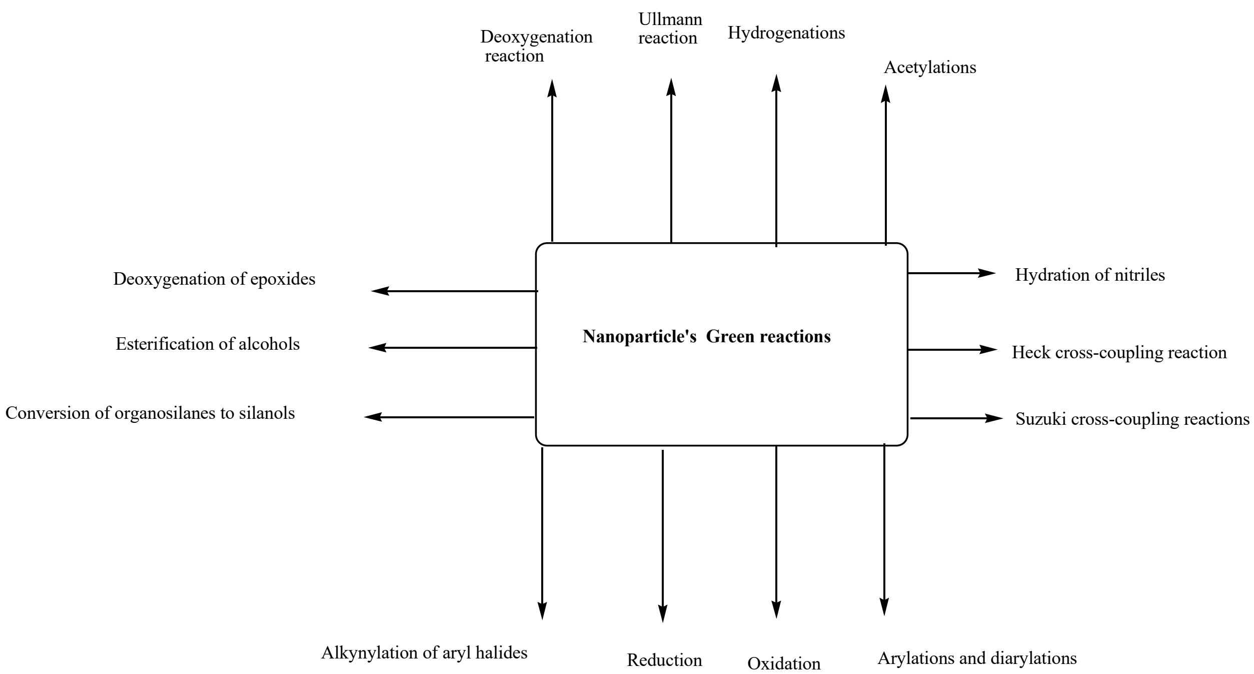


Figure 3: Varoius nanoparticles green reactions.

CONCLUSION

There been many different types of metal nanoparticles that have been used as catalysts for many reactions. In many cases, the metal nanoparticles are synthesized in aqueous solution in which water is the solvent, or is conducted in the presence of ionic liquids. There have also been cases where the nanoparticles are used as catalysts for different types of green reactions. Green reaction conditions include using water as the solvent, using solvent that is organic-free, conducting the reaction using ionic liquids, and running the reaction at atmospheric pressure. While there has been a lot of progress in applying the use of green chemistry to catalysis with nanoparticles, there is lot more room to further expand this field. In this review article, we have focused on the synthesis of various nanoparticles and their use in organic synthesis. Still there is need to explore and to synthesize new nanocatalysts with more properties. This review provides a comprehensive understanding on organic reactions which are catalyzed using environmentally friendly nanoparticles and nanocatalysts.

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CONFLICT OF INTEREST

No authors have stated any conflicts of interest.

CONTRIBUTIONS

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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