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# Recent advances of functionalized $Fe_3O_4$ as nanocatalyst in carbon-carbon coupling reaction: A review

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**Review Paper**

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**Abstract:**

Green Chemistry is the design of chemical products and processes that reduce or eliminate the use of hazardous substances. The benefits of green chemistry include safe food, clean water, clean air, less exposure to toxins, and safer consumer goods of all kinds. This approach is effective in safeguarding our ecosystem from harmful and poisonous materials. Scientists are becoming more interested in nanocatalysis since these processes are environmentally benign and safe. In the past ten years, magnetic nanoparticles have demonstrated remarkable efficacy as catalysts due to their ease of synthesis, modification, and enormous surface area ratio. Several of the most important features of these nanocatalysts are their high selectivity, excellent yields, and short reaction times, so these catalysts provide economical synthetic routes to target products. Organic reactions, including oxidation, hydrogenation, coupling, condensation, esterification, photocatalysis, and biocatalysts, ferrite nanocatalysts are coated with silica and are capable of catalyzing industrially. It is possible to synthesize transformations with excellent yields and selectivity using these nanocatalysts in a simple and eco-friendly manner. The catalytic activity of these catalysts can be reused for many repeated cycles as the magnetic properties enable easy separation after the reaction is complete by applying external magnetic fields. In this review, we focus on the synthesis of novel iron oxide nanoparticles using various synthetic methods. These catalysts are applicable in organic reactions such as Suzuki, Heck, Sonogashira, and $A_3$ coupling reactions. Also, we discuss the recyclability and re-utilization of these nanoparticles.

**Keywords:** Coupling reactions; Iron oxide nanocatalyst; Recyclability; Re-utilization; Silica-coated iron oxide

## 1. Introduction

For a few years, reagents, catalysts, and catalytic reactions have attracted the aim of finding meaningful advantages in the pharmaceutical and chemical industries. The development of the catalytic material will be required to push on material that can selectively catalyze the particular chemical reaction with a highly reactive, recycled through the simple separation method, and regenerate without any loss of catalytic properties. Applying the magnetic field on the nanoparticle to attract toward the external magnet and remove the magnetic field has been dispersed in a solution. The concept of green chemistry is to be a synthesis of the green catalyst, which is a part of sustainability. In the current issue, we discuss the synthesized nanoparticle supporting various metal oxides and ionic liquids with acid-functionalized compounds. Those catalysts are applicable to organic reactions and their recyclable study.

Green chemistry, also known as sustainable chemistry, is an ideology of chemical research and chemical engineering that inspires the design of products and activities that depreciate the use and generation of unusual substances. According to Green Chemistry, the reagents and catalysts are commercially available, inexpensive, and eco-friendly substrates that found huge applications in organic transformation [1–6]. Prevailing research on organic chemistry to the development of methodologies by using various green reagents, catalysts, and solvents (water and ionic liquid). The method is simple and eco-friendly to the environment as well as human health [7–9].

Synthesis of various organic reactions involves organic solvents that may be highly toxic, carcinogenic, and explosive, and the solvent is affected by the environment. Nowadays, increasing pollution causes a lot of issues that are brought forward at a global level. The oxidation reaction is for the synthesis of various organic compounds in the presence of solvents, reagents, catalysts, and synthetic methods. The recyclable green catalyst does not lose any catalytic properties. That catalyst gives an excellent yield of product and selectivity [10–17].

A few years ago, the development of the novel synthesis of ferrite magnetic nanoparticles was used in organic transformation. The magnetic nanoparticles are synthesized by changing the oxidation state of iron that occurs in Fe (II) and Fe (III), the catalyst giving magnetic properties. Frenkel and Doefman first reported on ferrite magnetic material in 1930, that ferrite magnetic nanoparticle size is about $10-100$ nm is conventional particle size, and below 10 nm particle size has super magnetic nanoparticles [18]. The nanoparticle was synthesized using various methods, such as co-precipitation, thermal decomposition reduction, and sol-gel [19–22]. The nanoparticle has been separated by using an external magnetic field.

## 2. Recent advance study of iron oxide nanoparticles

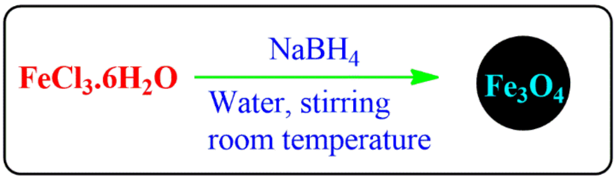
The silica-coated magnetic nanoparticle synthesis is done using the stobber method in the presence of tetraethyl orthosilicate, and it's separated by an external magnet. The nanoparticles were synthesized using tetraethyl orthosilicate. The nanoparticles were doping by using different metal oxides like $TiO_2$, Cu, Zn, Ni, Co, Cu, Pd, Pt, etc. The synthesized nanoparticle was characterized by using FT-IR, XRD, SEM, TEM, and EDS techniques that nanoparticle used in the organic reaction for synthesis. Physical properties of magnetic nanoparticles are the study of the smaller particles found to be less hazardous for the environment [23].

## 3. Synthesis of ferrite magnetic nanoparticles

There is a number of methods reported in the literature for preparing a spacious variation of ferrite magnetic nanoparticle some of which debate below. Which is an important inscription that experimentally and cost-efficacy of these processes directly affects the sustainability of the nanocatalyst.

### 3.1 Ferrite magnetic nanoparticles ($Fe_3O_4$)

The ferrite magnetic nanoparticle is synthesis by using various techniques and methods. S. H. Chaki et al. [24] reported a magnetic $Fe_3O_4$ nanoparticle synthesis by using a reduction technique (Scheme 1). In the presence of sodium borohydride ($NaBH_4$) and Ferric chloride hexahydrate solution under aqueous medium by stirring to obtain darker and black ppt. by Co-precipitation method. The magnetic nanoparticle has an average of $40-60$ nm. Ferrite magnetic nanoparticle was prepared as a reported method in the literature [25], and Nasr-Esfahani MASOUD et al. [26] prepared ferrite nanoparticles using $FeCl_3$ and $FeCl_2$ in an aqueous medium. The resulting solution was added dropwise into the aqueous base to form a black precipitate and remove the nanoparticle through a decantation process. The magnetic nanoparticle size is an average of $20-30$ nm. Swapnil R. Bankar et al. [27] reported the synthesis of ferrite nanoparticles by $FeCl_3.6H_2O$, urea, and $FeSO_4.H_2O$ and added aqueous base. Then, the mixture was ultra-sonicated at room temperature to obtain a black powder. The diameter of the nanoparticle ranged from 20 to 40 nm.

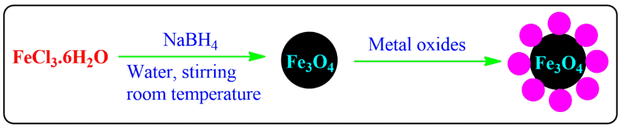


**Scheme 1.** Synthesis of ferrite magnetic nanoparticle.

### 3.2 Metal oxide ferrite nanoparticle ($Fe_3O_4$@Metal oxide)

Metal oxide supporting magnetic nanoparticles such as Au, Ru, Mo, Pt, and Pd are doping and utilized for organic transformation [28, 29]. N. F. Andrade Neto et al. [30] are reported to characterization and photo-catalytic applications of $Ce^{4+}$, $Co^{2+}$, $Mn^{2+}$, and $Ni^{2+}$ doped by using ferrite nanoparticle under the Co-precipitation method. D. A. Petrov et al. [31] reported the magnetic nanoparticle and magneto-optical properties of $Fe_3O_4$ nanoparticles modified with Ag. The study of the effect of Ag inclusion on magnetic properties and magnetic circular dichroism of nanoparticles in the mixing of ferrite and Ag nanoparticles. Y. Yang et al. [32] reported a facile fabrication of Au ferrite Nanocomposite as an excellent nanocatalyst for ultrafast recyclable nanocatalyst, which was synthesized by using a seed deposition method. Eman Alzahrani et al. [33] reported the photodegradation of Eosin Y using silver-doped magnetic nanoparticles. Which is most applicable in the purification of industrial waste from dyes. The nanoparticle was developed and synthesized using the co-precipitation method. Renpeng Yang et al. [34] reported an effect of Mg doping on magnetic induction heating of Zn-Co ferrite nanoparticles. Ferrite magnetic nanoparticles with the spinel structure are of great significance in magnetic induction hyperthermia. He has reported the influence of the $Mg^{2+}$ add on Curie temperature (Tc), Magnetic properties, and heating efficiency of $Mg_xZn_{0.8-x}Co_{0.2}Fe_3O_4$ ($0.1 \leq X \leq 0.5$) nanoparticle was synthesis by hydrothermal methods (Scheme 2).

Mona A. Mohamed et al. [35] reported to the cobalt ferrite magnetic nanoparticle as a highly efficient electrochemical platform for simultaneous determine the Dexlan-



**Scheme 2.** Synthesis of Metal oxide doping ferrite magnetic nanoparticle.

soprazole and Grainisetron hydrochloride. It was the first novel electrochemical sensor used in an experiment to determine the Dexlansoprazole and Granisetron hydrochloride. The nanoparticle was synthesis via the green hydrothermal method. Sami Ullah Rather et al. [36] reported determining the effect of Al doping in zinc ferrite nanoparticles, and their structural and magnetic properties were studied. Aluminum doped by zinc ferrite ($ZnFe_{2-x}Al_xO_4$) nanoparticle with x ranging from 0 to 0.9 was prepared by the thermal treatment method using a PVP capping agent. Kariim Ishaq et al. [37] reported to develop the nickel ferrite doped $\alpha$-alumina nanoparticle successfully and their characterization, as well as the antibacterial activities of the nanoparticle. The synthesized magnetic nanoparticle behaviors towards the biomedical applications of transition metal. The synthesis of alumina-based nickel-ferrite nanoparticles via wet impregnation method.

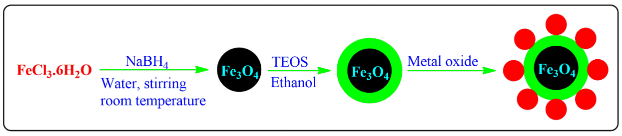
### 3.3 Ionic liquid supporting ferrite nanoparticle ($Fe_3O_4$@I.L)

Preet Shikha et al. [38] reported the synthesis of Mn doping-induced physic-chemical changes in Le-Ce ferrite nanofabricated by using an ionic liquid assisted hydrothermal route by supporting an ionic liquid surfactant. In this work, the effect of Mn substitution on the structural, optical surface, and magnetic properties of mixed $LaFeO_3$, i.e., La-Ce Nano ferrite synthesis via hydrothermal route by using ionic liquid. Yaping Zhang et al. [39] reported the synthesis of basic ionic liquid immobilized on magnetic nanoparticles using glycerol trioleate by transesterification. Ionic liquid supporting heterogeneous catalyst prepared by 1-allyl dodecyl imidazolium hydroxide basic ionic liquid immobilized by magnetic mesophore. Manika Dewan et al. [40] reported an efficient and reusable ionic liquid stabilized magnetic nanoparticle as a catalyst used in the aza and thia-Michael reactions.

### 3.4 Silica coated metal oxide nanoparticle ($Fe_3O_4$@$SiO_2$)

The ferrite magnetic nanoparticle was synthesized by using the Co-precipitation method and coated with the various ratios of $SiO_2$ as per the Stober method. Baskar Thangaraj, Zhao Hua Jio, Lingmei Dai, Dehua Liu, and Wei Du [41] reported an effect of silica-coated ferrite magnetic nanoparticle for lipase immobilization and applicable in bio-diesel product on the ferrite magnetic nanoparticle was synthesis by using the Co-precipitation method and coated by using TEOS presence of Stober method (Scheme 3) [42].

Suh Cem Pang, Sze Yun Kho, and Suk Fun Chin [43] reported on the fabrication of magnetite/silica/Titania core-shell nanoparticle synthesis by using the sol-gel method. In this study, $TiO_2$ nanoparticle show photo catalytic activities [44, 45]. previous work on $TiO_2$ ferrite magnetic nanoparticle reported by Li et al. [46] nanoparticle synthesis by using the Sol-gel method.



**Scheme 3.** Synthesis of silica-coated metal oxide nanoparticle.

## 4. Characterization of ferrite nanoparticles

Characterization of nanocatalyst was consummate in Transmission electron microscopy (TEM), time-of-flight-secondary ion mass spectrometry (TOF-SIMS), X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), inductively coupled plasma atomic emission spectroscopy (ICP-AES), and SEM elemental mapping with energy dispersive X-ray spectrometry (EDS). The technique that applies to the analysis of the chemical composition analysis of synthesized nanoparticles was done by energy dispersive X-ray analysis (EDAX) attached to XL 30 FESEM scanning electron microscopy. Determination of particle size and morphological study by using transmission electron microscopy (TEM) and scanning electron microscopy (SEM). The crystal structural study was done by using powder X-ray diffraction (XRD). The chemical bonding structure is determined by Fourier to transform Infrared spectroscopy (FT-IR). The optical absorption spectrum was observed on the spectrophotometer [47].

## 5. Applications of magnetic nanoparticles

In recent years, magnetic nanoparticle (MNPs) are attracting increasing interest in the various catalyst. The advantages of this catalyst are the development of a green and sustainable process in the organic transformation by using less toxic, solvent-free, or mild reaction conditions and recyclable catalysts. Methods give excellent yields of products and easily workup from the reaction [48–53]. A magnetically recyclable nanoparticle is used in coupling reactions, Oxidation-Reduction reactions, and multicomponent reaction synthesis [54, 55]. In the presence of nanocatalyst, the reaction goes through a green route. Catalyst has major applications in the selectivity of the reaction, easy separation, Higher TOF, and excellent stabilization. We provide a current review of the magnetic nanoparticle applications in the catalytic process and organic transformation.

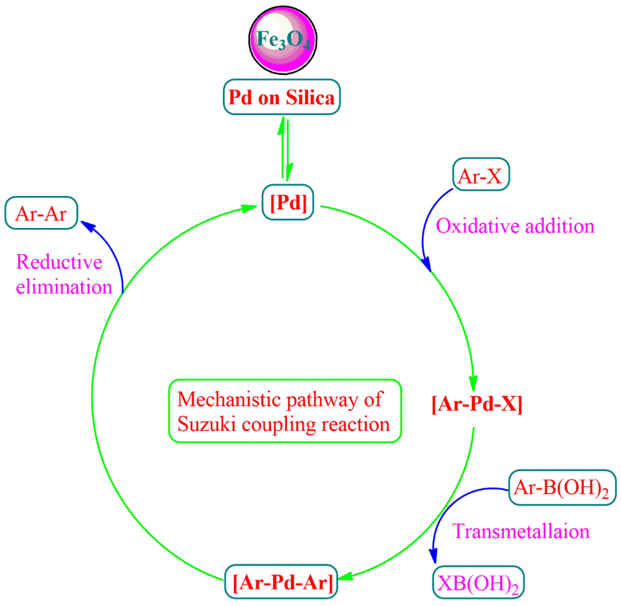
### 5.1 Coupling reaction or C-C bond formation

The disclosure of the chemistry of transition metal is the discovery in the 1960s and the change in the synthetic methods for organic transformation. Transition metal nanocatalyst has been expanding into excellent reagents in the coupling reaction. Palladium metal is a transition metal, which shows the reactivity and selectivity in the coupling reaction. Various Palladium-catalyzed reactions have come out in the past few years, and researchers have attracted and increased the number of publications in the area of research. There are two major impacts of an increase in the research area. The first is that this palladium metal catalyst easily forms the C-C bond and C-N bond formation under the effective reaction condition and simultaneous excellent yields of product. The reaction occurs in mild reaction conditions. Palladium-catalyzed numerous reactions, namely Suzuki, Sonogashira, Still, and Heck reactions carried out under mild reaction conditions, give the coupling product [56–58]. These reactions are carried out under palladium-catalyzed with various complexes [59, 60].

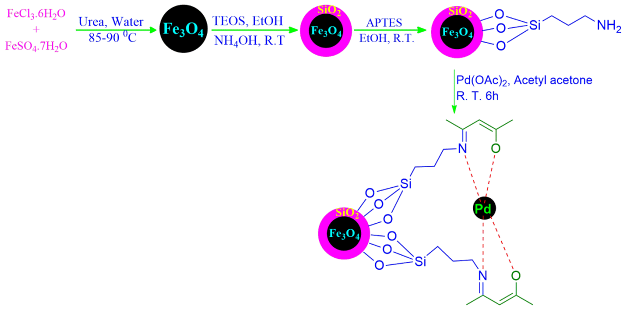
### 5.1.1 Suzuki coupling reaction

The Suzuki coupling reaction is most dominant in organic synthesis to evolve in the 20th century [61–64]. The Suzuki coupling reaction was first reported in 1979, which involves hydroboration of alkyne in the presence of catecholborane, followed under the Pd (0)-catalyzed organometallic complex. Suzuki reaction becomes one of the more effective for the synthetic route to prepare an arylation reaction. Those compounds are used to prepare natural products, pharmaceuticals, and industrial development [65–68]. The reason behind that is the first reason for Industrial development of the importance of this reaction condition to make it attractive to the synthesis of intermediates, molecules, and various drugs. The second reason the Suzuki coupling reaction is carried out in the presence of Organoborane is that it is easily available, highly stable, and less hazardous to the environment. Last third dominant reason Suzuki reaction work on the various method is useful in the organic synthesis. Bicyclic compounds join with coupling reactions which shows the biological activities, which is containing various active functional groups [69–72]. The typical catalytic cycle of the Suzuki coupling reaction involved postulated molecular, homogenous Pd catalysts that cycle between $Pd^0$ and $Pd^{II}$ oxidation state during catalytic reaction mechanistic pathway and similarly to silica supporting Pd catalyst also work (Scheme 4).

S. P. Vibhute et al. reported that the magnetic nanoparticle supported by Pd was catalyzed under the Suzuki coupling reaction. The design and synthesized a new magnetic nanocatalyst, a ferrite supporting silica coating amine-functionalized Schiff-Base-Pd (II) metal complex (Pd-AcAc-Am-$Fe_3O_4$@$SiO_2$) (Scheme 5) for the Suzuki-Miyaura cross-coupling reaction [73].



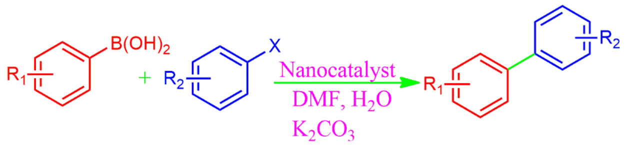
**Scheme 4.** Mechanistic pathway of Suzuki coupling reaction.



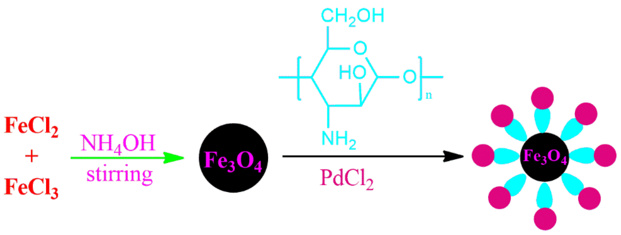
**Scheme 5.** Synthetic route for the preparation of nanoparticle (Pd-AcAc-Am-$Fe_3O_4$@$SiO_2$).

Suzuki Miyaura cross-coupling reaction is carried out in the presence of homogenous and heterogeneous Schiff bases, which gives excellent yield. The homogenous catalyst is not easily separated from the reaction mixture, but the heterogeneous catalyst is easily separated out and again recycled from the reaction, then it will be reused in the next reaction [74–77]. Therefore, author was synthesis the silica-supported ferrite magnetic nanoparticle with Schiff base and used to synthesis of Suzuki-Miyaura cross-coupling reaction. The mixture of aryl halides ad aryl boronic acid in aqueous DMF with $K_2CO_3$ mild basic condition and a catalytic amount of nanocatalyst then the mixture was reflux at 80 °C, in this reaction study of various solvents namely as toluene, ethanol, acetonitrile, THF, Dioxane water, and DMF. But a reaction is carried out in the presence of aqueous DMF which gives the 98% Yield of product. The nanocatalyst provided nearly quantitative yield for electron-rich and electron-deficient aryl halides contain the iodine or bromine substituent (Scheme 6). The recycling of nanocatalyst was performed by magnetic concentration, washing with ethanol, and dry well without any purification. Without loss of catalytic properties was observed after several reaction cycles. The nanocatalyst system was shown to be more rapid than an analogous solid-phase system. The author believes that the increased rate of reaction is due to the surface-active site.

A. Naghipour and A. Fakhir et al. reported a heterogeneous $Fe_3O_4$@chitsan-Schiff base supporting Pd nanocatalyst used for the Suzuki coupling reaction. The design of a magnetic nanoparticle-supported palladium catalyst via a 2-pyridine carboxaldehyde complex on the surface of a ferrite magnetic nanoparticle (Scheme 7) was used in the Suzuki coupling and also Heck coupling reaction [78]. Chitson is a naturally occurring polysaccharide, which is a biologically active material that can be easily available and



**Scheme 6.** Synthesis of Diarylation of Suzuki-Miyaura cross-coupling reaction.

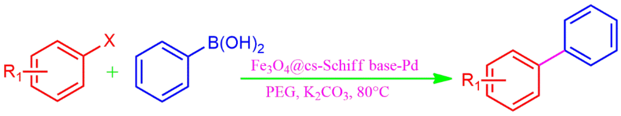


**Scheme 7.** Synthesis of $Fe_3O_4$CS-Schiff base-Pd nanocatalyst.

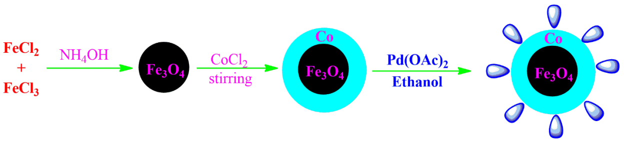
less harmful material [79–82]. In this method to develop a greener heterogeneous catalyst was developed by magnetic core-shell type $Fe_3O_4$CS-Schiff base-Pd nanoparticle used in the coupling reaction. Using $Fe_3O_4$CS-Schiff base-Pd Nano catalyzed Suzuki-Cross coupling reaction between the aromatic halide (Br, I) and phenylboronic acid dissolved in the polyethylene glycol (PEG) and mild base $K_2CO_3$ was achieved in satisfactory yields. This catalyst could be recycled and reused three times by using different aromatic halides and phenylboronic acid. In this reaction also study the Electron donating and electron-withdrawing substituent gives an excellent yield of product (Scheme 8).

K. K. Senapati et al. was reported to the synthesis of Pd nanoparticle supported on cobalt ferrite nanoparticle achieved by the addition Pd-nanoparticle during the synthesis of cobalt ferrite nanoparticle by Ultra-Sound irradiation by Co-precipitation method. The Suzuki coupling reaction in ethanol under ligand-free condition. In this reaction low catalyst was loaded (1.6 mol %) and separated by using an external magnet and then reused for a number of the cycle with sustained catalytic activity (Scheme 9) [83].

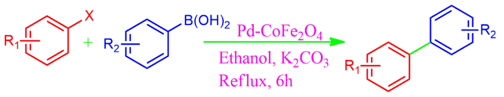
The reaction between aryl halide with phenylboronic acid under the catalytic amount of $CoFe_2O_4$ magnetic nanoparticle and other some ligand, Schiff bases attached to Pd-catalyzed reaction [84] in the presence of $Na_2CO_3$ mild basic condition. But the author's reaction is carried out in the presence of Pd-$CoFe_2O_4$ magnetic nanoparticle was used in the coupling reaction. The recyclable study of magnetic nanocatalyst was also examined by using repeated runs on the same batch of the catalyst in the next Suzuki reaction. This catalyst which recycles four times without loss of catalytic activities. Catalyst is easily separated by using an external magnet and washing with ethanol and then again used (Scheme 10).



**Scheme 8.** Suzuki coupling reaction by using $Fe_3O_4$CS-Schiff base-Pd nanocatalyst.



**Scheme 9.** Synthesis of Pd-$CoFe_2O_4$ magnetic nanoparticle.

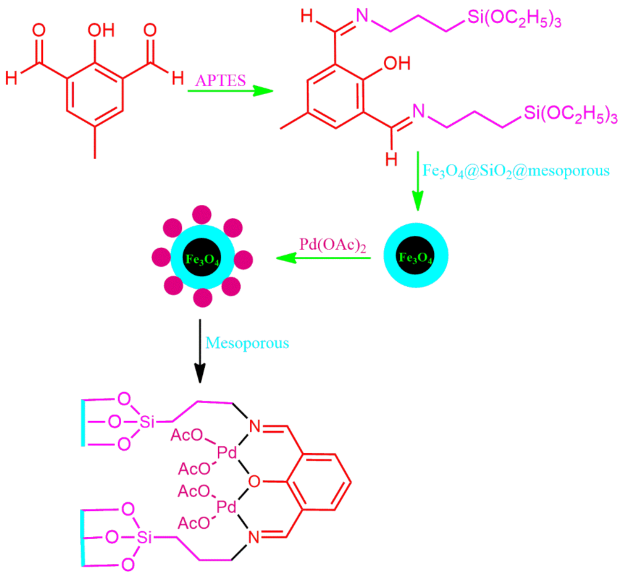


**Scheme 10.** Suzuki coupling reaction under Pd-$CoFe_2O_4$ catalyzed nanoparticle.

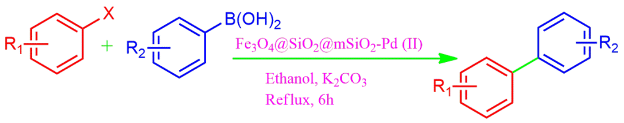
Zhengping Dong and Jiantai Ma as reported to the synthesis of the Suzuki-Miyaura Cross-coupling reaction catalyzed by efficient and recyclable $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (II). The Pd (II) complex functionalized core-shell magnetic mesoporous catalyst was synthesized by APTES under ultrasound irradiation. The $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (II) catalyzed was synthesis by using APTES was dissolved in toluene and reflux 105 °C under nitrogen for 6 h then added mesophore nanocatalyst and $Pd(OAc)_2$ then ultra-sonication for 30 min (Scheme 11) [85].

Suzuki coupling reactions were performed by aryl halide, phenylboronic acid, $K_2CO_3$ mild base in ethanol, and $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (II) nanoparticle mixture was stirring at 80 °C to gives coupling product. Magnetic mesoporous silica nanoparticle-supporting Pd (II) catalyst was stable, reusable, and recovered by external magnet. The catalyst was recycled by 6 time without loss of catalytic activities (Scheme 12).

W. Li et al. is reported to the preparation and



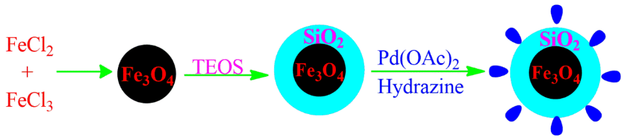
**Scheme 11.** Synthesis of core-shell $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (II) nanoparticle.



**Scheme 12.** Suzuki coupling reaction by core-shell $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (II) nanoparticle.

characterization of the novel immobilized nanoparticle $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (0) with large pore-size mesoporous for Suzuki coupling reaction (Scheme 13). Core-shell magnetic mesoporous $Fe_3O_4$@$SiO_2$@m$SiO_2$ microsphere with double shell structure which is prepared from coated by silica layer. The inner shell of the magnetic core highly effective. The porous outer shell can provide more surface area and add the amount of palladium on the surface of the porous shell. The catalyst which having $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (II) is reduced by using sodium borohydride we get $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (0). The material was then tested as a mesoporous catalyst in the Suzuki coupling reaction, which is carried out in the presence of aryl chloride, phenylboronic acid, and mesoporous $Fe_3O_4$@$SiO_2$@m$SiO_2$-$NH_2$@Pd (0) nanocatalyst in mild basic condition, water as a solvent. The reaction gets the expected product with excellent yield after 6 h at 80 °C (Scheme 14). The author indicates that after completion of the reaction catalysts will be drying and reused at least six times without any significant loss of activity. The material was dispersed in both aqueous and organic solvents [86].

Ardeshir Khazaei et al. are reported to the synthesis of palladium supporting $Fe_3O_4$@$SiO_2$ nanoparticle used in the Suzuki coupling reaction. The $Fe_3O_4$@$SiO_2$-Pd nanoparticle was synthesis by using waste eggshell as a natural base. $SiO_2$ nanocatalyst was synthesized by using simply from risk husk biomass as the source of bio silica. $Fe_3O_4$@$SiO_2$-Pd used in Suzuki coupling reaction in the presence of waste egg which is easily available, Inexpensive, and simple experimental procedure. The eggshell nanoparticle which synthesis by silica under reflux condition then adds palladium we get $Fe_3O_4$@$SiO_2$-Pd (II). The reduction of this catalyst by using hydrazine to obtained $Fe_3O_4$@$SiO_2$-Pd (0) reactive nanocatalyst (Scheme 15).



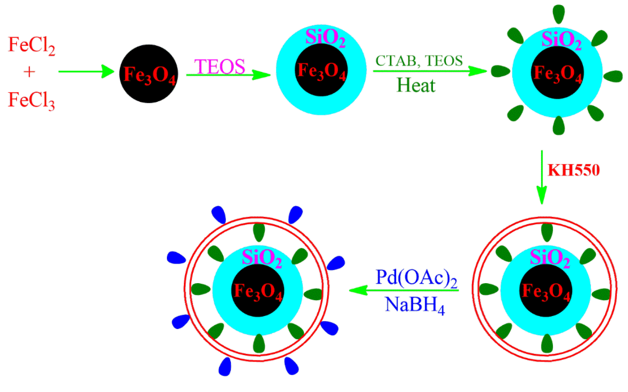
**Scheme 15.** Schematic illustrated for preparing $Fe_3O_4$@$SiO_2$-Pd (II).

The coupling reaction between iodobenzene with phenylboronic acid was carried out using $Fe_3O_4$@$SiO_2$-Pd (0) under aqueous ethanol under the solvent at 85 °C to gives the coupling product (Scheme 16). The optimization study the effect of electron-withdrawing and electron-donating functional group in aryl iodide and bromide on the catalytic activity we get corresponding coupling product [87].

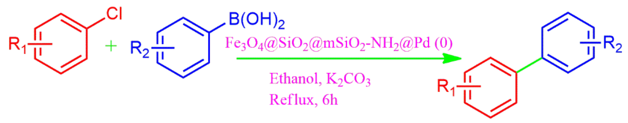
### 5.1.2 Heck coupling reaction

The Heck coupling reaction is the most important is used for the formation C-C bond (Scheme 17). Depending upon the coupling partner of an alkene with aryl halide which allows the arylation, alkylation, or vinylation of a various alkene by corresponding aryl halides [88–90]. Heck cross-coupling reaction is more applicable in organic synthesis and the pharmaceutical industry in the last two decades Nobel Prize winner to Heck, Suzuki, and Negishi in 2010. Heck coupling reaction undergoes Pd-catalyzed under mild basic condition. It involved the molecular homogenous catalyst that cycle between Pd (0) and Pd (II) oxidation state during the catalytic reaction. The various ligand and Pd catalyst which is co-ordination with various polymer and ionic liquid [91, 92].

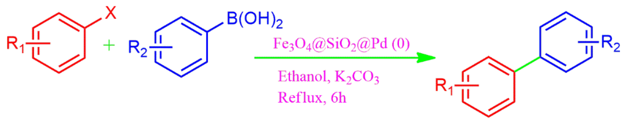
S. P. Vibhute et al. reported the synthesis of palladium supporting Schiff-base complex immobilized on magnetic nanoparticles used for Mizoroki and Matsuda Heck coupling reaction. The synthesis of Pd-AcAc-Am-



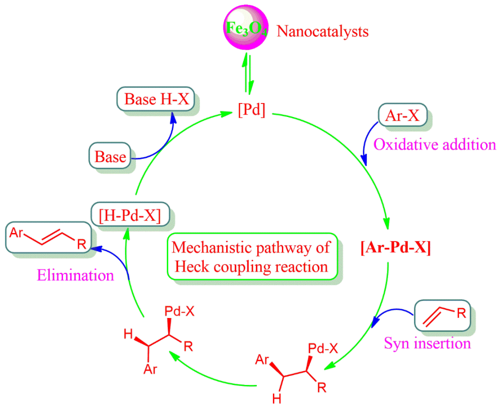
**Scheme 13.** Schematic illustrated for preparing immobilized $Fe_3O_4$@$SiO_2$@m$SiO_2$-$NH_2$@Pd (0).



**Scheme 14.** Suzuki reaction under catalyzed by $Fe_3O_4$@$SiO_2$@m$SiO_2$-Pd (0).



**Scheme 16.** Suzuki-coupling reaction by $Fe_3O_4$@$SiO_2$-Pd (0).

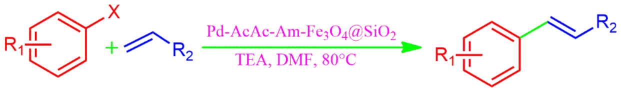


**Scheme 17.** Mechanistic Pathway of Heck Coupling Reaction.

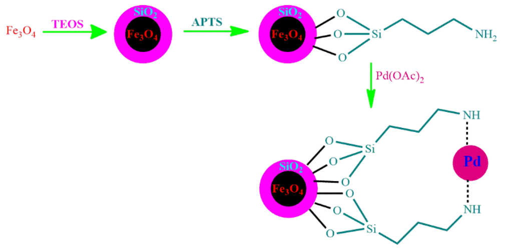
$Fe_3O_4$@$SiO_2$ nanoparticle as shown in (Scheme 5). Synthesized Pd-AcAc-Am-$Fe_3O_4$@$SiO_2$ nanocatalyst was tested for Heck reaction in the presence of aromatic halide, Vinylic substrates like acrylate, styrene, and acrylonitrile under triethylamine as a base in DMF reflux at 80 °C for 5 h to give coupling product (Scheme 18) [93].

Z. Wang et al. reported the synthesis of Palladium supporting magnetic nanoparticles catalyzed by the Heck coupling reaction (Scheme 19). The palladium-supporting nanoparticle was synthesized by the mixture of $H_2PdCl_4$ solution, dissolved in ethanol, and then the PVP mixture was refluxed for 3 h. Isolating APTs coated $SiO_2/Fe_3O_4$ nanoparticle from the reactant, the prepared solution of Pd nanoparticle was added and stirring for 3 h. The magnetic nanoparticle was synthesized through wet impregnation in the corresponding palladium nanoparticle and ferrite nanoparticle in the $H_2PdCl_4$ solution. The catalyst showed high activity and could be recovered and reused several times. The cross-coupling of acrylic acid with aromatic halide dissolved in acetonitrile solution added $Fe_3O_4$@$SiO_2$-Pd (0) catalytic amount then added sodium acetate solution was prepared in water, and the mixture was refluxed at 80 °C for 12 h will get the coupling product (Scheme 20) [94].

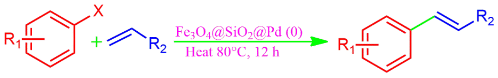
A. S. Singh et al. reported that the synthesis of palladium supporting on zinc ferrite nanoparticle is used in a Heck coupling reaction. Superparamagnetic solid catalyst has been synthesized by loading Pd (0) during the synthesis of zinc ferrite nanoparticle by ultra-sound assisted Co-precipitation in the surface stabilized or capping agent. (Scheme 21) The scope of this catalyst for the Heck reaction was studied for a wide variety of aryl halides with various alkene. The catalyst separation and recyclability of the catalyst were estimated using separation, and the recyclability of the catalyst was estimated using the Heck coupling reaction. The catalyst was recycled 5 times with no loss of catalytic properties [95].



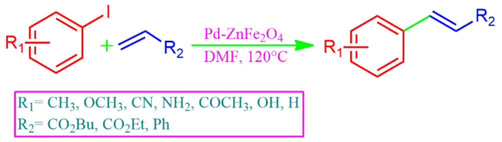
**Scheme 18.** Heck coupling reaction catalyzed by Pd-AcAc-Am-$Fe_3O_4$@$SiO_2$.



**Scheme 19.** Schematic representation of $Fe_3O_4$@$SiO_2$-Pd (0).



**Scheme 20.** Heck coupling reaction catalyzed by $Fe_3O_4$@$SiO_2$-Pd.



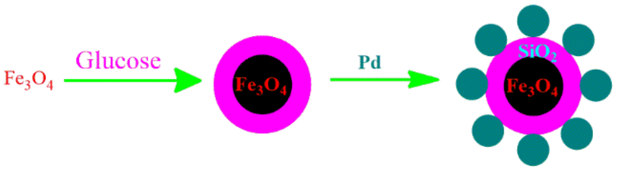
**Scheme 21.** Heck coupling reaction of Pd-$ZnFe_2O_4$.

Maiyong Zhu and Guowang Diao reported that the synthesis and characterization of magnetic carbon nanocomposite palladium supported immobilized magnetic $Fe_3O_4$@C nanoparticles has been prepared by a three-step process in the report. The author also studies the morphology, inner structure, and magnetic properties of all products. Nanocatalyst, which is used in the Heck coupling reaction (Scheme 22).

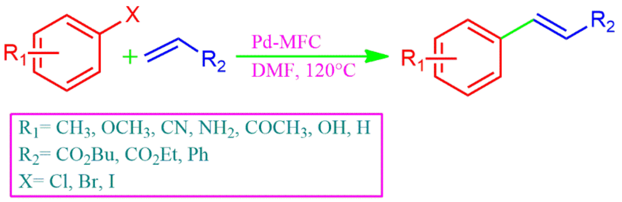
The $Fe_3O_4$@C composite was synthesized by the carbonation of glucose in the presence of $Fe_3O_4$ nanoparticle prepared by the hydrothermal method [96]. The $Fe_3O_4$ nanoparticle is dispersed in the water which contains glucose and ultra-sound irradiation. Then transfer of reaction in an autoclave and kept at 200 °C for 12 h. The nanoparticle was collected by using an external magnet. Finally drying nanoparticle in a vacuum for 24 h at 60 °C (Scheme 23) [97].

### 5.1.3 Sonogashira coupling reaction

The coupling of any terminal alkynes derivatives in the presence of palladium (0) catalyzed reaction which gives the coupling product [98]. The reaction is performed by using a base like triethylamine, and the reaction generally uses copper iodide as a catalyst. The reaction goes through mild conditions, usually at room temperature, which means the reaction occurred with thermal sensitivity [99–105]. The substituted acetylene is the main advantage of designing various biomolecules, naturally occurring compounds, and in pharmaceutical industries. [106–108] Some other methods are less virtual because the number of disadvantages like haemolytic coupling takes place and the formation of other



**Scheme 22.** Schematic representation of Pd/MFC.

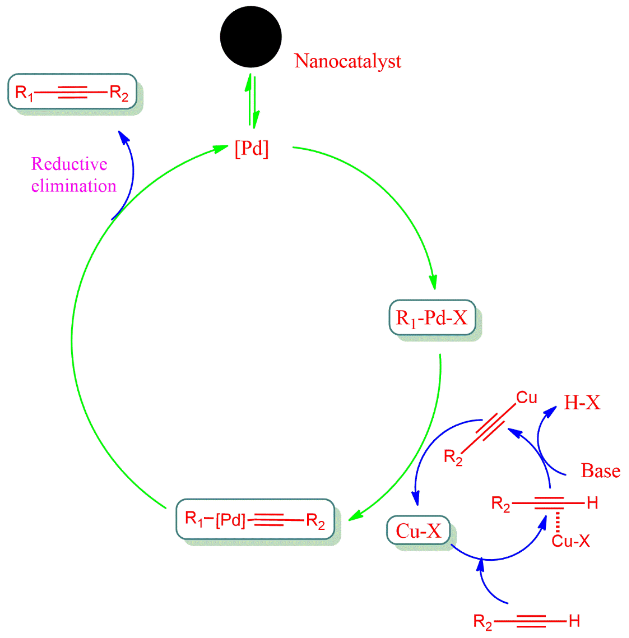


**Scheme 23.** Heck reaction catalyzed by Pd/MFC.

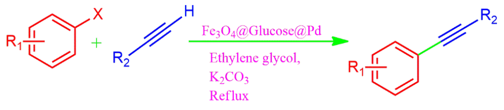
products but the Sonogashira coupling reaction in the presence of aromatic alkyne gives the specific coupling product [109–112]. The mechanistic path of the Sonogashira coupling reaction as shown (Scheme 24).

Habib Firouzabadi and Nasser Iranpour reported a ferrite magnetic nanoparticle catalyzed by the Sonogashira-Hagihara coupling reaction in the presence of ethylene glycol using the Ligand condition [113]. Sonogashira-Hagihara coupling reaction is an important invention in organic synthesis. In this method, the various metal-supporting (Pd, Ni, Cu, and Co) magnetic nanoparticle was studied [114]. The reaction of aromatic halide with aryl acetylene molecule with the ferrite nanoparticle in the aqueous base like $K_2CO_3$ and ethylene glycol reflux at 125 °C for appropriate time will get coupling product (Scheme 25). This method is highly effective because the nanoparticle is easily separated by using an external magnet, and ethylene glycol is water-soluble and will separate by using the distillation method, both recycled and reused for the next reaction.

J. M. Nagarkar et al. reported to the synthesis of palladium on manganese supported magnetic nanoparticle used in the Sonogashira coupling reaction. The author is reported that the synthesis of diary acetylene by using phenyl propionic acid and propiolic acid with the arene diazonium salt catalyzed by Pd-$MnFe_2O_4$, which was synthesized by using Ultra-Sound assisted under the Co-precipitation method [115]. The coupling reaction of propionic acid and arene diazonium salt was carried out under mild basic conditions like $K_2CO_3$ and solvent DMF in the presence of a nanocatalyst. We get the coupling product (Scheme 26). We have developed a green and reusable, magnetically separable Pd$MnFe_2O_4$ nanocatalyst for the decarboxylative Sonogashira reaction of phenyl propiolic acid with the diazonium salts, which could be prepared from inexpensive. Various diazonium salts are converted into symmetric and unsymmetrical aryl acetylenes derivatives in moderate to excellent yield. Due to the study of the comparative study between Pd NPs and $MnFe_2O_4$ NPs, the catalyst could be reused and recycled without any loss in catalytic activity.



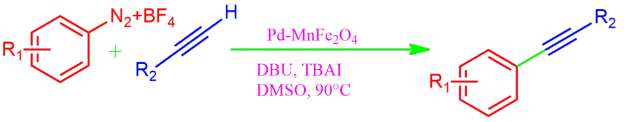
**Scheme 24.** Mechanistic pathway of Sonogashira coupling reaction.



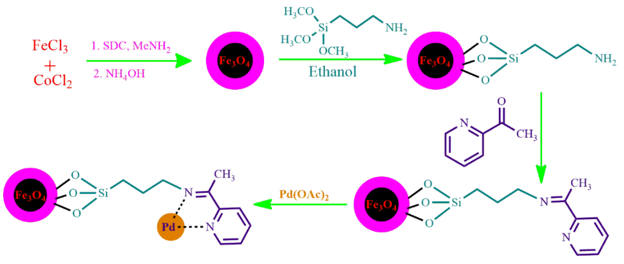
**Scheme 25.** Nano ferrite catalyzed under Sonogashira coupling reaction.

N. T. S. Phan and H. V. Le reported to the synthesis of super magnetic nanoparticle supported phosphine free palladium catalyst used for the Sonogashira coupling reaction [115]. Authors to develop the super magnetic nanoparticle by using a functionalized Schiff base which on the surface of the immobilized ligand. The amino-functionalized magnetic nanoparticle dispersed in ethanol and added 2-acetyl pyridine. The resulting mixture was sonicated for 30 min and then refluxed with stirring for 36 h. The nanoparticle was collected through an external magnet and dry well under vacuum. Immobilized Schiff base, palladium acetate dissolved in acetone, and nanoparticles were sonicated for 30 min at room temperature. After completion of the reaction collect the nanoparticle by using a magnet and dry it well (Scheme 27).

A mixture of iodobenzene, phenylacetylene, phenylacetylene, $K_3PO_4$, and n-hexadecane as the internal standard in dimethylformamide in the presence of the amount of immobilized palladium-catalyzed and copper (I) iodide catalyst. The reaction mixture was stirring under the immobilized palladium catalyst was favoured for both the electron-withdrawing group and the electron-donating group to slow down the cross-coupling process (Scheme 28). Recovery of catalyst from the reaction by using an external magnet. The scope of this catalyst for the Sonogashira coupling reaction was studied for a wide variety of aryl halides with various acetylene. The catalyst separation and recyclability of the catalyst were estimated using the separation and the recycla-



**Scheme 26.** Sonogashira coupling reaction under Pd-$MnFe_2O_4$ nanoparticle.



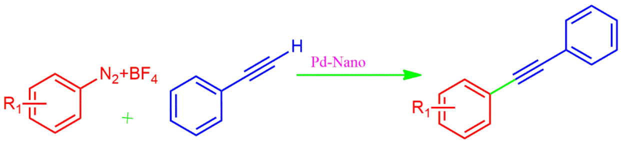
**Scheme 27.** Synthesis of Pd catalyzed immobilized super magnetic nanoparticle.

bility of the catalyst was estimated using the Sonogashira coupling reaction. The catalyst was recycled 5 times no loss of catalytic properties.

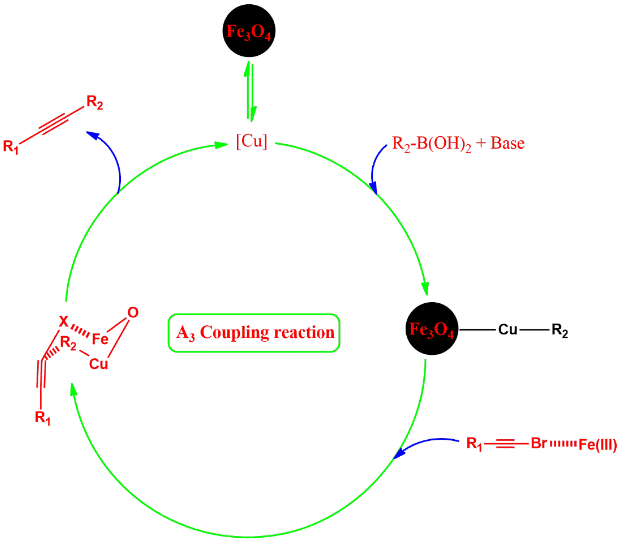
### 5.1.4 $A_3$ coupling reaction

$A_3$ coupling is the multicomponent reaction that gives the Propargylic amine derived from aldehyde, alkyne, and amine coupling reaction. The reaction was first reported by Chao-Jun Li [104, 116]. The reaction between aldehyde alkyne and amine gives Propargylamine, which shows the biological activity. Therefore, Propargylamine derivatives are used in the chemical industry and pharmaceutical industry [105–107]. In recent years, propargylamine synthesis is reported as a coupling catalyst prepared from Nobel metals (Co, Ag, Au, and Ir). But this metal is an expensive and hazardous catalyst; therefore, it develops the recyclable ferrite nanoparticle used in the $A_3$ coupling reaction. The reaction of aldehyde, alkyne, and amine in the presence of a nanocatalyst mechanistic path as shown in (Scheme 29).

Gonghua Song and Chao-Jun Li reported to the synthesis of propargylamine using the ferrite recoverable nanoparticle. The reaction between aromatic aldehyde, alkyne, and amines was developed $A_3$ coupling in the presence of recyclable nanoparticle [108, 117]. The reaction goes through a green route, an economical, practical, and efficient process. The reaction between aromatic aldehyde, sec. amine and phenylacetylene in the presence of iron oxide nanoparticles under the appropriate solvent to give coupling products. After completion of the reaction, the nanoparticles separated by using a magnetic field and again recycled. In this reaction, a good yield was obtained, and the catalyst was reused 12 times without any need for activation (Scheme 30).



**Scheme 28.** Sonogashira coupling under super magnetic nanoparticle.

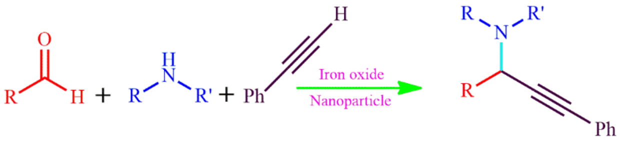


**Scheme 29.** Mechanistic pathway of $A_3$ coupling reaction.

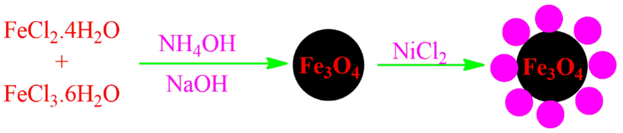
Alinezhad et al. reported a multicomponent $A_3$ coupling reaction catalyzed by biosynthesized magnetic $Fe_3O_4$@Ni nanoparticle from Euphorbia Maculata extract. In this work, biosynthesized $Fe_3O_4$@Ni nanoparticle is used to synthesize 2, 3 di-substituted benzo [b] furan using the aldehyde, secondary amine, and acetylene derivatives. The author was developed which are the current green, practical, and economical methods in minimum reaction time and give an excellent yield of the product. The Ni-supporting ferrite magnetic nanoparticle was prepared using $FeCl_3.6H_2O$ and $FeCl_2.4H_2O$ dissolved in an aqueous extract of Euphorbia Maculata aerial part reflux at 70 °C under vigorous stirring to change the color and make it darken. Then the addition of $Na_2CO_3$ 1 M solution and maintaining pH is about 10. After completion of the reaction collect the nanoparticle by using an external magnet and wash with water several times. The nanoparticle was kept at 60 °C for 12 h to dry it well (Scheme 31). The mixture of an alkyne, secondary amine, and aldehyde under $Fe_3O_4$@Ni nanoparticle was stirring ethanol in atm $N_2$ for the appropriate time to gives the coupling product propargylamine derivatives (Scheme 32). After completion of the reaction, the catalyst was to recycle and reuse to next for six-time in the model reaction [109, 118].

## 6. Future scope

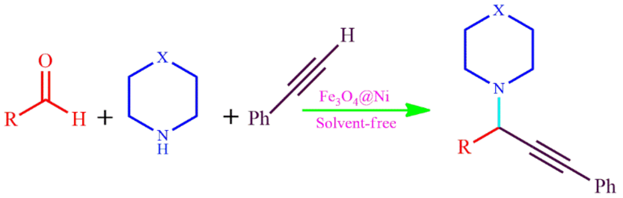
We aim to current a broad research area on magnetically recyclable nanocatalysts. The important thing about this work is the use of ferrite magnetic nanomaterial as a green catalyst. We can be talking about the need for green chemistry aspects that are eco-friendly to the environment. The green



**Scheme 30.** Synthesis of Propargylamine by using ferrite nanoparticle.



**Scheme 31.** Synthesis of $Fe_3O_4$@Ni nanoparticle used for the $A_3$ coupling reaction.



**Scheme 32.** $Fe_3O_4$@Ni nanoparticle catalyzed under Multicomponent $A_3$ coupling reaction.

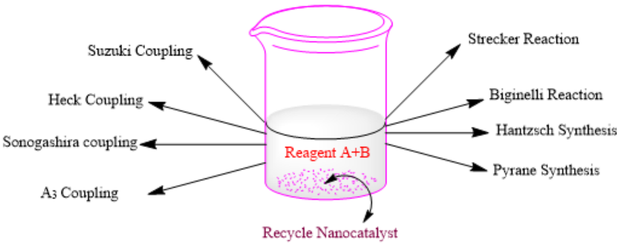


**Figure 1.** Scope of the nanoparticle.

reagent is inexpensive, easy to available, recyclable, and gives excellent yields under solvent-free conditions. The reagents easily recycle after completion of the reaction without loss of catalytic properties. The focus on environmental pollution increases in the presence of toxic chemicals, catalysts, and solvents used for the organic transformation to development and design in the synthetic methods used in the reaction and control of the pollution (Figure 1).

We then discuss the key focus of this review, which is a study of reactions and transformations using ferrite magnetic nanoparticles. We start with the synthesis of ferrite nanoparticle and silica-coated ferrite magnetic nanoparticle using various synthetic methods with supporting metal-based (Pd, Pt, Ru, Rh, Au,) and ionic liquid supported catalysts. Discuss the application of these catalysts for synthetically important carbon-carbon coupling reactions, such as Suzuki, Heck, Sonogashira, and $A_3$ coupling reactions. Finally, we review the current research area of magnetic nanomaterials used for environmental applications, in particular for an eco-friendly catalyst. And the ending of this review focuses on the applications of nanomaterials is in the synthetic methods that give the excellent yield of product and selectivity of product.

## 7. Concluding remark and outlook

In this review, we have a detailed study on greener methods and techniques used for the synthesis of ferrite magnetic nanoparticle supported by using green extract, ionic liquid, metal oxides, and acid functionalized compound under simple or ultra-sound irradiation technique. There is increasing research interest in the use of green or eco-friendly materials, which is applicable in the various synthetic methods. There has been an increasing tendency of the application of magnetically recyclable material to evolve more systematic and green methods. This review has provided a large-scale overview of the application of ferrite nanoparticle used in the synthesis of coupling reactions (Suzuki, Heck, Sonogashira, and $A_3$ coupling). The focus on nanocatalyst recyclable nanoparticle was used in the reaction and it was again reused study.

**Authors contributions**
Not applicable

**Availability of data and materials**
The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Conflict of interests**
The author declares that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

**Open access**

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