RECENT ADVANCE ROLE OF Fe3O4-SUPORTED NANOCATALYSTS IN VARIOUS ORGANIC TRANSFORMATIONS

Abstract

The importance of magnetic nanoparticles as to provide the large surface area during catalytic reaction. It show dynamic change in catalyst activity when a solid support material which have magnetic property. Such type of solids called quashi-homogeneous catalyst easy extraction and removal from reaction mixture using an external magnet and such catalyst can be reused. They show environmentally benign green catalytic process. For synthesis of magnetically isolable nanoparticles has been different methods such as i) Wet chemical, ii) Templet directed, iii) Micro emulsion, iv) Thermal decomposition, v) Solvothermal method, vi) Solid state deposition method, vii) Spray pyrolysis, viii) Self-assembly method physical and lithographic technique etc. Several screening strategies to deliver them and suitable for catalytic applications such as polymer, silica and carbon coating of magnetic nanoparticles reported in literature. This review focused on significant progress for prevarication of nano structural catalyst with unique emphasis on screening and fictionalization of magnetic nanoparticles. Accounting the importance of coupling reaction chemistry in organic transformations wide applications of magnetic nanoparticles based catalyst in several type of coupling reactions and various other organic transformations.

Keywords: Nanocrystals, nanoparticles, Organic transformation

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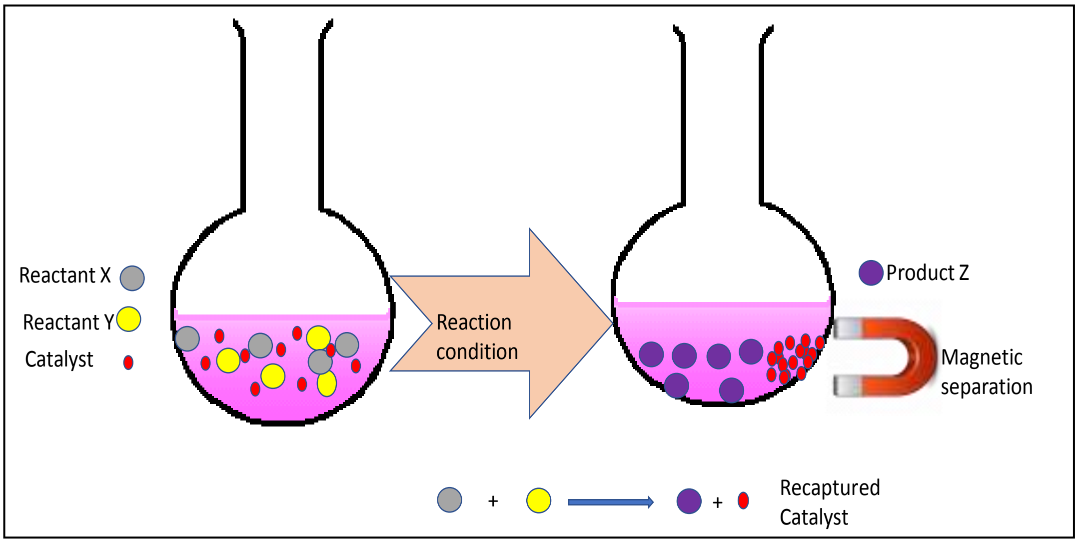


MNPs Supported Catalyst Separation

I. INTRODUCTION

Nanoparticles play salient property in the research area as modern technology due to wide novel applications. They have remarkable properties and large surface area to volume ratio. They are intermediate between atomic and bulk levels and due to this reason, the wide research has been done for synthesis various morphologies for nanoparticles like 1-dimensional,nanotubes,nanorods,nanorings,nanowires,nanosheets,nanoflowers,nanosphere etc.

Recently nanomaterials with solid support have been gives widely applicable in the designing various heterogeneous catalysts[30-34]. To resolve economic and environmental issues, eliminates filtration, centrifugation, energy consumption, catalyst lost, and time for recovery of catalyst.

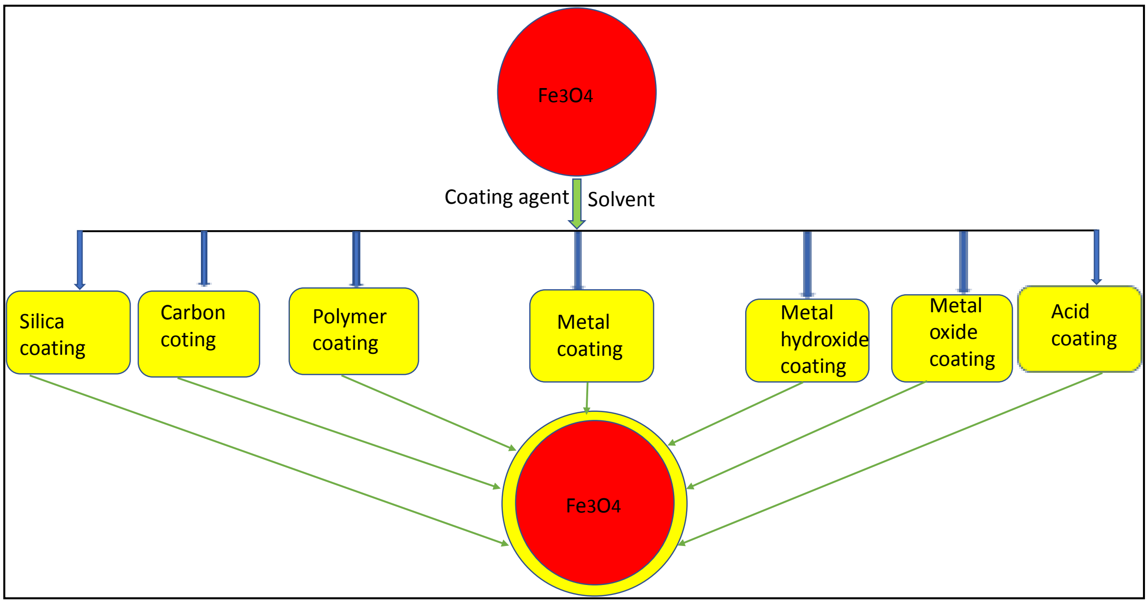
Magnetic nanocatalysts have wide range of applications involving in the catalysis, magnetic fluid recording, biotechnology, biomedicine, material science, photo catalysis, electrochemical and biomaterial sensing, microwave absorption, magnetic resonance imaging, medical diagnosis, data storage, environmental remediation and electrode for super capacitors [35-46]. Recently super paramagnetic properties of solid supported nanoparticles has wide applications but those are highly sensitive to oxidation and reduction. Hence to protect solid nanoparticles from oxidation and reduction the screening of MNPs is must be workout. The perspective of this review provides wide overview about planning and growth of magnetically recoverable nanocatalysts.

The planning for protection against oxidation, reduction, and corrosion is provided after the synthetic approaches for magnetic nanoparticles synthesis. Additionally described are their uses as supported catalysts on magnetic nanoparticles in diverse organic transformations.

II. MODERN ADVANCES IN THE MANUFACTURING OF MAGNETIC NANOPARTICLES

The synthetic route for designing of magnetic nanoparticles plays important role in controlling the particle framework, it is constitution, magnetic and surface characteristics which effectively enlarge their applications in wide variety of multidimensional field. Many research review focused on synthesis of high quality magnetic nanoparticles by different methods like Co-precipitation, Thermal decomposition, Micro emulsion, Solvothermal, Hydrothermal process, Template-mediated synthesis use of ultrasonic irradiation, Microwave reactor, Lamer approach for controlled synthesis[47,48].The MNPs have synthesized by using above mentioned methods are chemically inert, has a high surface area to volume ratio, and is thermally stable, but because of magnetic nature there is agglomeration of particles takes place and it affect the size as well as properties of MNPs,So in this review we generally give different methods to protect the nanoparticles and its use in organic transformations.

To avoid agglomeration, oxidation ,reduction, corrosion of MNPs different strategies are used as guard to protect the nanoparticles surface by various coating agents like silica, carbon, polymer, metal, metal hydroxide, metal oxide and acid coating[49].



Coating policies embedded for regulating MNPs

III. IMPORTANCE OF CATALYSTS IN VARIOUS ORGANIC TRANSFORMATIONS

Magnetic nanoparticles captivated entire organic chemistry as probably useful catalyst through their magnificent characteristics. Examining the increasing attention on approving the greener pathway for organic transformations involving conversion of all reactant into product with minimization of energy as well as waste and good atom efficiency.

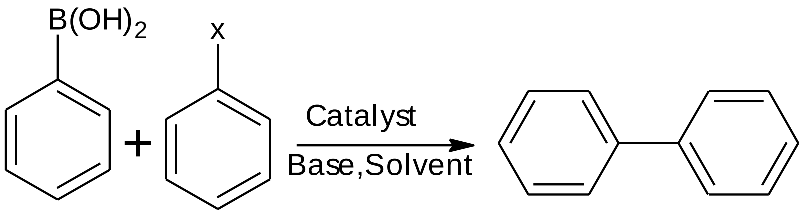
Here we explain a) coupling reactions and b) multicomponent reactions. Coupling reactions further classified into homo coupling (C-C) and hetero coupling (C-N,C-S, C-O,C-This review focus the utilization of Fe3O4 supported catalysts in various organic transformations. The spontaneity of separation of catalyst by using magnet from reaction medium and reusability are the important qualities of catalytic system based on magnetic nano support.

IV. C-C COUPLING

The formation of C-C bond through cross-coupling reaction represents as one of the most powerful tool in various organic transformations and have effective features in organic synthesis as well as pharmaceuticals, agrochemicals, material science, fine chemicals [50-53]. Transition metal catalyst have been designed as excellent reagent for synthesis of carbon-carbon simple and asymmetric reaction. The most significant carbon-carbon cross coupling processes include the Suzuki, Heck, Sonogashira, Stille, and Hiyama coupling reactions, which are often catalysed by homogeneous catalyst.

To perform the coupling reactions greener number of challenges are again for effective separation and sequential recovery of homogeneous catalyst, the developed MNPs supported transition metal catalyst becomes effective alternative for these reactions.

V. SUZUKI COUPLING REACTION



The Suzuki coupling reaction or Suzuki–Miyaura reaction couples an aryl halide to an aryl boronic acid to form a variety of biaryl derivatives which shows significant components for various medicines, natural products, and polymers. Various factors are taken into account for the development of this reaction both academic and industrial level. For Suzuki coupling reactions, Pinhua Li and colleagues employ magnetic nanoparticles supported palladium as a highly reusable catalyst [54] they use phosphine functionalized magnetic nanoparticles with good control on catalyst size, reusability of catalyst is about eight times with efficient yield. The MNPs are formed by simple co-precipitation method.

Momhad G. and co-workers design magnetic nanoparticles supported oxime palladacycle catalyst [55].They use this type of catalyst because lot of work is done by using oxime palladium complex as a homogeneous catalyst but there is lack of recyclability and to avoid this development done is use of various supporting medium like Polystyrene, resins, silica, mesoporous organo-silica but magnetic supported oxime palladacycles is excellent one to reusability as well as catalyst activity.

Xuanduong le and co-workers use magnetic mesophorous silica nanoparticles-supported Pd(II) [56]catalyst for C-C Suzuki coupling reactions. Magnetic mesoporous silica support is more favourable for the mass transfer to reactant molecule to their short pore channels, and it was simple to separate from the reaction mixture.. Catalyst is moisture and air stable are reusable and give good yield of product up to six cycles and it gives potential applications in industrial synthesis.

Yu long and co-workers design palladium immobilized on dopamine functionalized magnetic nanoparticles[57]. Dopamine generally used as surface modification of unfunctionalized Fe3O4 NPs because of them solvent dispensability and facile templet free synthetic route. Instead of employing conventional linkers, dopamine with an amino group was employed as a surfactant and an interparticle linker. It also improve the solvable property of NPs in aromatic solution by using this catalyst we can prepare carbonyl ative coupled product with excellent yield and good TOF.

HojatVeisi, and co-workers design biologically and environmentally benign chitosan-biguanidine supported MNPs. Chitosan contains hydroxy and amino group which coated with magnetite and functionalization is carried out by using guanidine and finally formation of complex with palladium[58]. Catalyst shows excellent role in coupling reaction with efficient reusability.

Palladium nanoparticles supported on reduced magnetite graphene oxide were developed by S. Jafer and colleagues as nanocatalysts[59], for such coupling reaction. Because of using reduced graphene oxide as well as palladium NPs here is large surface area and due to this it gives excellent catalytic activity as well as yield of product at room temp in efficient time.

Graphene is a layer of sp2 hybridised carbon that is one atom thick and densely packed into a two-dimensional honeycomb lattice. It serves as the fundamental building block for all other graphitic materials [60]. Recently, a variety of metal oxide nanoparticles (NPs), including Fe3O4, Co3O4, TiO2, and ZnO [61-64] may be impregnating on graphene oxide (GO) sheets to provide this material additional functionality because of its expansive surface area.

Through a click reaction, Tahereh Azadbakht created a water-reliable palladium complex that was deposited on magnetite, coated with silica, and then linked with imine functionalized silane and (2-[2-(2-formylphenoxy) ethoxy] benzaldehyde) ,hydroxyaryl palladium complexes immobilised on magnetite nanoparticles[65].The catalyst may be collected, reused, and readily separated for further reactions.

For the stabilization of palladium nanoparticles and its use as a catalyst in Suzuki-Miyaura coupling reaction in aqueous media under mild and low palladium loading conditions, Mohammad Gholinejad and coworkers [66] designed palladium supported phosphonite functionalized MNPs phosphinite-functionalized magnetic nanoparticles with imidazolium ionic liquid moiety.

Ionic liquids (ILs) are categorised as "green solvents" because they have unique characteristics including non-volatility, non-flammability, and a wide temperature range across the liquid phase. However, the majority of ionic liquids are high-cost substances, making it undesirable economically to employ them as solvents. Nevertheless, by immobilizing ILs on the surface of solid support, organic processes can happen on the surface of the support's thin IL layer while reducing the amount of ILs that are used.

S-benzylisothiourea combination of palladium support and modified Fe3O4 magnetic nanoparticles (Pd-SBTU@Fe3O4) as a stable in both moisture and air catalyst was developed by Arash Ghorbani [67]. Catalyst could be reused up to five times without any significant loss of its activity.

Arash Ghorbani□Choghamaranidevlopeisatonic anhydride functionalized magnetic supported catalyst the ring opening of anhydride by amine group of Fe3O4@APTMS**[68]** gives amino benzoic acid linked with Fe3O4@APTMS and it forms complex with palladium acetate to give Pd(0)-ABA- Fe3O4 catalyst, which is useful for c-c coupling reaction. It results in higher product yields with reusability of catalyst up to six cycles within efficient time.

The possibility of using Schiff base transition metal complexes as catalysts in a variety of processes has led to substantial research into these compounds.[69,70] These complexes have been widely employed in a variety of chemical processes, including the hydrogenation of organic substrates, [71] the epoxidation of olefins,[72] the conversion of epoxides into halohydrines,[73,74] the asymmetric ring opening of terminal epoxides, and oxidation reactions[76,77]. Therefore, Hassan Keypour and colleagues continue to modify support-tethered amino groups with Schiff base ligands chemically by forming the appropriate imines by a reaction between amine and aldehyde groups of 4-hydroxy-3-methoxybenzaldehyde. In the crosscoupling reaction between aryl halides and phenylboronic acid, they also investigated the catalytic behaviour of a Pd(II) complex of the tethered Schiff base ligand[78]. Reusability and the production of products with good yields in a timely manner are key functions of a catalyst.

Elaheh Farzadrecently devlope new protocol Fe3O4/SiO_2@PDA/Pd [79] as a magnetically separable catalyst for Suzuki coupling reaction as well as some reductions. A biomolecule called dopamine is made up of several catechol and amine groups. By forming relatively stable linkers between the hydroxyl groups on their surfaces and the appropriate anchoring agents, such as phosphonic acid and dopamine derivatives, the metal oxide nanoparticles can spontaneously functionalize to coat a thin layer of surface-adherent polydopamine (PDA) on various material surfaces.[80]PDA, which has a high concentration of phenolic hydroxyl and amine functional groups, can be used to coat metal oxide nanoparticles (MNPs) to change their surface. They verified that it is possible for polyphenols or catechols from PDA to combine with polyvalent cations in aqueous solutions, which can enhance the surface characteristics and ability of Fe3O4@PDANPs to adsorb and decrease metal ions. The $[Na_2PdCl_4]^{2+}$ ions, the synthesised palladium precursor, may be readily absorbed on the PDA surface to decrease metallic palladium.

Neda Seyedi and collaborators used natural resources to synthesise Pd NPs, which they then complexed with grafted imines to make modified Schiff bases on graphene oxide[81]. The schiff base complexes are good options for creating nanoparticles because they have distinctive chemical and optical features. The catalytic activity of the prepared catalyst was investigated by employing this coupling reaction. Plant extracts have been suggested as a useful alternative to chemical approaches for the manufacture of Pd nanoparticles in order to overcome the previous drawbacks. Several articles on the production of palladium nanoparticles (Pd-NPs) from plants, including Soybean (Glycine max) leaf extract[82], Cinnamomum zeylanicumbark[83], C. Camphoraleaf[84], and Curcuma longa tuber[85], are known.

The Pd-dithizone complex designed by Arash Ghorbani-Choghamarani [86], supported by MNPs, and used in the Suzuki-Miyaura coupling reaction of phenylboronic acid with various aryl halides. With great reusability and reaction completion in a very short amount of time, this catalyst may be easily retrieved from the reaction mixture by using an external magnet..

Recently, Fatemeh Heidari and colleagues created a magnetically separable Fe_3O_4@SiO_2/isoniazide/Pd nano-catalyst [87] for the Suzuki coupling reaction's synthesis of bi aryls. Here, just isoniazide is used as a novel functionalizing group to create heterogeneous catalysts [88-91] that are safe for the environment and carry out the reaction..

In this case, silica nanoparticles are used to prevent the agglomeration of magnetite produced from rice husk, an agricultural waste product, according to Ardeshir Khazaei's newly developed green methodology for the manufacture of amino functionalized magnetically separable palladium nanocatalyst [92-93]. It has developed into a source for the production of silicon compounds, including silicon nitride, silicon carbide, and silicon dioxide[94].The fabrication of nano Fe_3O_4@SiO_2-Pd employed SiO_2 nanoparticles that had been produced. Because it contains important minerals and amino acids, eggshell is a widely available natural food item that is consumed around the world. The majority of eggshell trash is often disposed of without any prior preparation. Eggshell is entirely recyclable, biocompatible, and biodegradable[95]. In addition to organic materials and water, eggshells include a network of protein fibres and crystals of calcium carbonate, calcium phosphate, and magnesium carbonate. More than 90% of them are made of $CaCO_3$[96].In this study, CaO derived from eggshell was used as natural solid support for Suzuki coupling reaction. The used catalyst shows excellent reusability and environmental benign, reaction carry out within short time interval

In order to produce flexible GO-based nanocomposites, the many oxygen-containing functionalities available on the GO surface may properly act as coordination sites for dispersing metal nanoparticles. Simply said, just graphene-based catalysts without any functionalized joint groups can exhibit the same efficacy and stability, if not superior. In this study, Wenzhi Fu and colleagues described a simple, environmentally friendly, and cost-effective method for depositing palladium nanoparticles on magnetic rGO nanosheets without the use of surfactants or functionalized joint groups. The extensive surface area of rGO[97] was evenly dispersed with Pd and Fe_3O_4 nanoparticles. The synthesised nanocomposites exhibit remarkable multifunctional catalytic activity in Suzuki-Miyaura coupling processes, providing improved yield and good reusability in short periods of time.

Magnetic nanoparticles functionalised with amines allow us to make a Schiff base and improve catalytic properties. Hossein Khojasteh and co-workers recently develop new protocol for magnetically supported catalyst functionalized with APTES and finally formation of schiff bases with 2-pyridine carbaldehyde or 2-thiophene carbaldehyde and again complex formation with palladium to form highly efficient magnetically supported catalyst [98].Here reusability of catalyst is better in presence of thiophene Schiff base complex with better yield of product.

Chitin undergoes alkaline deacetylation to produce chitosan (CS), the second most prevalent natural polysaccharide after cellulose.Because free amine and hydroxyl groups are present in CS, it can be subjected to many chemical modifications.[99] One of its adaptations is the construction of the Schiff base with CS.For the Suzuki and Heck reactions, [100] described heterogeneous catalysts based on chitosan.[101] The development of heterogeneous catalysts for organic transformations based on chitosan has been of interest to us for a few years.[102]

Here Anuradha and co-workers usemagnetic NPs coated with chitosan and it functionalized with Schiff base by using vanilinefinally they use palladium acetate to form catalyst**.[103]** The formed catalyst is magnetically separable, reusable up to five cycle without decreasing efficiency of product.

Recent research by Shima Asadi and colleagues has led to the development of a catalytic system known as Pd/GO/ Fe_3O_4/PAMPS that includes Pd-NPs immobilised on the surface of graphene oxide (GO) modified by poly 2-acrylamido-2-methyl-1-propansulfonicacid (PAMPS), and embellished with magnetic Fe_3O_4 [104]. In the Suzuki cross coupling process, this catalyst was successfully employed. The designed catalyst gives excellent reusability with better yield up to seven cycleswithout loss of appreciable catalytic activity within short time interval.

To boost the Pd catalyst's ability to recycle and further enhance its catalytic activity. Design of a magnetically separable sporopollenin-based Pd(II) catalyst by Talat Baran and colleagues. For the first time, they were able to remove the sporopollenin microcapsules from J. cinerea. afterwards the addition of magnetite particles, we used an amine functionalization procedure called silylation, followed by Schiff base formation to create coordination sites for the palladium ion. Microcapsules made of sporopollenin loaded with Pd(II) were put to the test in the microwave-assisted synthesis of biaryl compounds[105].

Fe_3O_4 magnetic nanoparticles functionalized by triazole are shown by Arefeh Dadras and colleagues as an effective anchor for the chelation of palladium. Suzuki-Miyaura C-C cross-coupling reactions have been successfully conducted using this catalyst. Fe_3O_4@SiO_2@3glycidoxypropyltrimethoxysilanetriazole[106] has been proved to be an effective chelating agent for copper ions, producing a durable catalyst for click reactions with little leaching and excellent reusability. Using phenylacetylene, sodium azide, and copper chloride as catalysts, silica-coated Fe_3O_4 magnetic nanoparticles were functionalized[107]. Copper was then removed from the catalyst by reacting with KCN, and palladium was added by reacting with $PdCl_2$ and KCl.Catalyst may be recycled up to 10 times without suffering significantly from reduced reactivity. Another advantage of this catalyst is its resistance to oxygen, moisture, and heat.

According to Hamid Mostafavi and colleagues, a Pd(0) complex covalently bonded with 2-formylbenzoic acid and supported by iron oxide nanoparticles treated with TEOS and APTMS can serve as a catalyst for the Suzuki coupling process, which produces biphenyls.[108]

Tannic acid, a chemical that resembles humic and has phenolic hydroxyl and carbonyl functional groups, can change the surface of Fe3O4NPs. Additionally, by complexing with them in aqueous solutions, it can enhance the Fe3O4's surface characteristics and capacity for adsorption and reduction of palladium ions.

The use of green solvents like water, non-toxic and safe ingredients, economic effectiveness, and appropriateness for pharmaceutical applications are just a few of the numerous benefits that come with bioinspired catalyst synthesis.[109] Such synthetic methods also don't need a lot of energy, heat, or pressure. Despite the availability of inexpensive technologies, the use of biological plant extracts for the synthesis of NPs has not yet been substantially investigated.[110,111]Tannic acid is one of the biological components that may be employed in the manufacture of NPs. It is a plant polyphenol that may be extracted from various foods and beverages, including pears, grapes, sorghum, bananas, lentils, black-eyed peas, and chocolate, in addition to other condensed tannins. Other beverages that include green tea, black tea, coffee, red wine, and beer.[112,113]

Similar to many other polyphenols, it exhibits anti-oxidant[114-117], anti-mutagenic[118-119], anti-carcinogenic, and anti-mutagenic effects.In order to create a new magnetically separable and reusable catalyst, Hojat Veisi and colleagues have designed Fe3O4@TA NPs[120] that have the ability to operate as both a reducing and a stabilising agent in the immobilisation of Pd NPs.

Fatemeh Rafiee and co-workers design silica coated NPs functionalized with N-Amidinoglycine amino acid, and its activity in suzuki cross-coupling reactions. The prepared Fe3O4@SiO2@*N*-amidinoglycine@Pd(0) **[121]**catalyst shows good reactivity, recyclability and time efficiency,and there is large scope to use various amino acids for functionalization.

In order to explore its activity in the Suzuki cross coupling process, Fatemeh Rafiee and colleagues reported on the synthesis and characterisation of a palladium catalyst based on Fe3O4@SiO2functionalized with vitamin B1 [122]. A high magnetic and outstanding dispersibility palladium nanocatalyst based on Fe3O4@SiO2 functionalized with thiamine hydrochloride (VB1) was created. With the support of a magnet, the catalyst may be easily removed from the reaction system and recovered. It can then be reused several times without significantly losing its effectiveness. Some distinctive characteristics of this heterogeneous magnetic NHC-Pd complex include the catalyst's stability towards air and humidity, utilisation of a minimal load of this environmentally beneficial palladium catalyst, ease of handling, and recyclability.

Pectin-carboxymethyl cellulose composite (Pct-CMC) with magnetically separable Pd nanoparticles was developed recently by Talat Baran in the catalytic processes. The catalyst Pd NPs@Pct-CMC/Fe3O4 [123]demonstrates outstanding catalytic activity against Suzuki-Miyaura reactions and exhibits good efficiency and reusability.

The Fe3O4/Ethyl-CN material, developed by Bahareh Abbas Khakiani and colleagues,[124] might be a useful support material for metal ions.For the Suzuki coupling reaction in a green medium (H2O-EtOH), they provide an easy method for synthesizing a recoverable Fe3O4/Ethyl-CN/Pd nanocatalyst. Fe3O4/Ethyl-CN/Pd nanocomposite, the created catalyst, has good magnetic properties and can be separated from the reaction mixture using a magnet. It has promise for use in industry since it can be recycled seven times without losing its catalytic activity.

Recently, HojatVeisi and colleagues designed an in situ method for immobilizing Pd nanoparticles on the surfaces of magnetic nanoparticles (MNPs) that are protected from view by a layer of green tea extract.[125]This method eliminates the need for hazardous and damaging reducing agents. Biphenyls are prepared by ultrasonicating the Suzuki-Miyaura reaction in water with the synthesised catalyst NPs.The generated Pd/Fe3O4 NPs exhibit great activity and dispersibility in water/ethanol (1:1), while being affected by ultrasonic waves, and without the use of a magnet.The catalysts may be recycled and reused up to seven times before losing their effectiveness, and they can yield more output in less time.

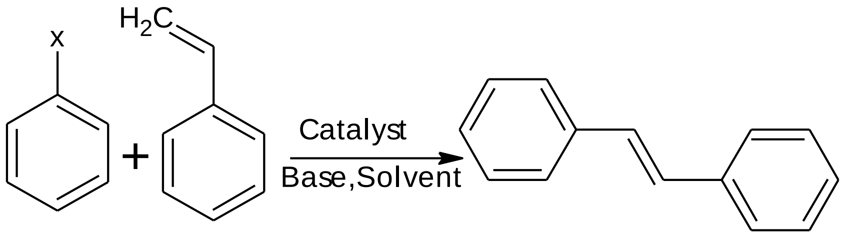
Najmeh Amirmahani and co-workers design Fe3O4@SiO2-APTMS-SAL-Pd [126] as novel catalyst for Suzuki reaction catalyst have good recyclability in efficient time and reaction also gives good yield.

Palladium complexed with 2-(7-amino-4-methyl2-oxo-2H-chromen-3-yl)acetic acid modified in magnetic nanoparticles (Pd@Fe3O4/AMOCAA) was developed by TaiebehTamoradi and colleagues to catalyse the traditional Suzuki and Sonogashira cross-coupling reactions[127]. The most crucial characteristics of this reaction are its high yield, rapid reaction, catalyst separation, and capacity to be repeated.

Ali Maleki and co-workers design o-phenylenediamine-functionalized Fe3O4/SiO2[128] magnetic nanoparticles for Suzuki reaction the prepared catalyst have good recyclability and good reaction yield.

Pradeep M. Mhaldar and co-workers design and synthesize nanomagnetic catalyst, a magnetite supported silica protected amine functionalized Schiff base–palladium(II) complex (Pd-AcAc-AmFe3O4@SiO2)[129]

VI. HECK REACTION



The Heck reaction, which is one of the most effective and significant reactions for the production of carbon–carbon bonds, causes a variety of alkenes to be arylated, alkylated, or vinylated by coupling with aryl halides. [130-132].

In order to create imidazolinium salt on magnetic nanoparticles for use in the Heck Reaction, Agnieszka Z. Wilczewska and colleagues designed the MNP@NHC-Pd complex[133]. Excellent recycling and effectiveness may be seen in the generated catalyst.

The metal complexes of phosphonates and phosphinites, which are significant phosphorus-based ligands in organometallic chemistry, have been employed in several catalytic processes [134], because phosphine ligands are frequently moisture and air sensitive, which causes problems in the catalytic system[135]. Mohsen Esmaeilpour and colleagues report the use of polymer-imid-Pd functionalized Fe3O4@SiO2 nanoparticles (Fe3-O4@SiO2-polymer-imid-Pd)[136] in absence of phosphine ligands.

As a novel magnetically recyclable heterogeneous catalyst in the Mizoroki-Heck reaction, Hojat Veisi and colleagues reported the production of palladium nanoparticles that were added into Fe3O4/diaminoglyoxime nanocomposite (Fe3O4/DAG/Pd)[137]. Palladium immobilized on Fe3O4/DAG nanocomposite, which they examined, was found to be an efficient nanocatalyst in the Mizoroki-Heck reaction of aryl halides with styrene, especially for less reactive aryl chlorides. Its advantages included ease of preparation, quick separating after the reaction using an external magnet, and multiple reuses with no noticeably losing catalytic activity.

Hassan Keypour and colleagues use Schiff base ligands to chemically modify surface-tethered amino groups, which are created when the amine and aldehyde groups of 4-hydroxy-3-methoxybenzaldehyde combine. Additionally, they looked at the Heck cross coupling reaction's Pd(II) complex of the tethered Schiff base ligand as a heterogeneous catalyst[138]. Reusability and the production of products with good yields in a timely manner are key functions of a catalyst.

Schiff bases are refers to a class of potent organic ligands that are frequently used in transition metal-catalyzed processes.[139,140] A novel class of Schiff base called iminomyridine bidentate ligands have emerged as desirable catalysts for Pd-catalyzed polymerization[141], which cyclization[142], or Heck processes[143].For the Heck reaction of aryl halides with olefins, Qiang Zhang and colleagues used a silicon-coated nano-Fe3O4-supported iminopyridine Pd complex (Pd(OAc)2@MNP[144].

The magnetic nanoparticle-N-heterocyclic carbene-palladium complex, or MNPs-NHC-Pd(I) [145] complex, was developed by Abdol R. Hajipoura and colleagues. It was used in the Mizoroki-Heck cross-coupling processes.With the use of a strong and recyclable catalyst, this method is very efficient and produces high yields of products in quick reaction times and under friendly circumstances.

ArashGhorbani- Choghamaranidevelopisotonic anhydride functionalized magnetic supported catalyst, the ring opening of anhydride by amine group of Fe3O4 @APTMS gives amino benzoic acid linked with Fe3O4 @APTMS and it forms complex with palladium acetate to give Pd(0)-ABA-Fe3O4 [146]catalyst, which is useful for c-c cross coupling reaction. .It gives better yield of product with reusability of catalyst up to six cycles within efficient time.

It is important to note that the shape and number of surface (vertex, edge, and plane) atoms of NPs also affect their catalytic activity because different crystallographic facets in various shapes can produce various reactivities and selectivities. In a study titled Pd cNPs/C@Fe3O4[147], Basuvaraj Suresh Kumar and colleagues investigated the catalytic activity of cubical Pd nanoparticles with surface facets combined with a magnetic support

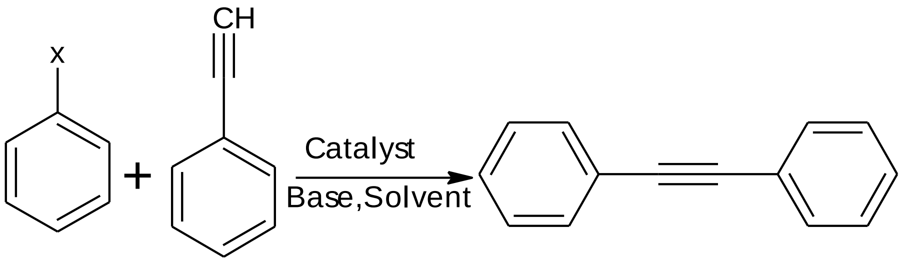
(C@Fe3O4) for Mizoroki-Heck coupling reactions.Additionally, they contrasted the shape-dependent catalytic activity of Pd cNPs with that of Pd sNPs for Mizoroki-Heck coupling processes.

Tetrazole chemistry has become widely used by researchers due to its significance, particularly in the disciplines of organometallic and pharmaceutical materials chemistry. With significant atom efficiency, noteworthy yields, and little byproducts, these reactions offer strong regioselectivity to produce 1H-tetrazoles. An efficient and broadly applicable method for the manufacture of tetrazole-functionalized nanomagnetic Fe3O4-supported palladium[148] nano-particles and their use in Heck coupling process is reported by Ali Elhampour and colleagues.

In order to create a hybrid nanostructure of Pd-Fe3O4 [149], Fangchen Zhen and colleagues have developed a one pot hydrothermal approach. Heck reactions were effectively catalysed by the Pd-Fe3O4, which also exhibited favourable magnetic characteristics and solvent dispersibility. Low temperature experiments showed that the Pd-Fe3O4 catalyst had outstanding catalytic activity. Easy preparation, excellent reactivity, and appropriate durability are just a few benefits that this catalyst provides. Additionally, the Fe3O4 component of the catalyst's magnetic qualities make it possible for the catalyst to be readily separated from its magnetic component and recycled, enhancing the catalyst's economic value and making it suitable for use in large-scale industrial applications. To increase the Pd-Fe3O4 hybrid nanocatalysts' capacity to be recycled, future study will concentrate on preventing Pd from leaching during the reaction.

A heterogeneous, magnetically recoverable, and reusable palladium catalyst is created by Marulasiddeshwara M. Bharamanagowda and colleagues and supported by a brand-new "hybrid coreshell" (Fe3O4-Lignin@Pd-NPs) [150]. With the help of n-butyl acrylate and styrene, the novel catalyst has been used in the Mizoroki-Heck reaction of different aryl/heterocyclic halides..

VII. SONAGASHIRA COUPLING



The sonogashira coupling reaction, which is commonly mentioned in organic chemistry for the synthesis of conjugated compounds, provides an excellent method to generate C-C bonds by combining aryl halides with alkynes[151-153].

Under heterogeneous ligand-free conditions in ethylene glycol, Habib Firouzabadi and colleagues demonstrate the effectiveness of magnetite (Fe3O4) [154] as a catalyst for the production of carbon-carbon bonds. Aryl iodides and activated heteroaryl bromides react with alkynes to form arylalkynes when this catalyst is used. An external magnetic field makes it simple to separate the catalyst from the reaction mixture. Without noticeably losing much of its catalytic activity, the separated catalyst can be recycled for a number of subsequent cycles.

For the Sonogashira-Hagihara coupling of aryl halides and phenylacetylene without the use of copper or phosphine, Mohsen Esmaeilpour and colleagues created Schiff base complexes[155] of metalions immobilised on superparamagnetic Fe3O4 nanoparticles and looked into their use.

Sonogashira-Hagihara coupling reactions with ligands and without copper can be catalysed by the same catalyst several times without noticeably losing its catalytic activity. The major benefits of this catalyst are its ease of use, great yields, rapid reaction times, heterogeneous nature, simplicity of separation, and recycleability.

CuFe2O4 nanoparticles supported on spherical silica were recently developed by Mohammad Gholinejad and colleagues [156], and palladium nanoparticles were then assembled. The novel magnetically recoverable catalyst has been effectively used in Sonogashira coupling reactions of aryl iodides and bromides with alkynes at 50oC under phosphine-free reaction conditions.

Wei Li and colleagues develop Fe3O4/SiO_2/P(GMA-co-EGDMA) is a novel magnetic material [157]. Hyperbranched/linear polyethyleneimine ligands were used to create composite nanoparticles. Through complexation between Pd2+ ions and multifunctional organic ligands, nano palladium was successfully anchored on this carrier, resulting in a new supported Pd nanoparticle catalyst with good dispersion, reusability, and high product yield.

The Sonogashira reaction is typically conducted in an organic solvent like an amine, benzene, THF, or DMF with a complex palladium catalyst working with CuX (X = Cl, Br, I) as a co-catalyst under inert conditions, which makes it hazardous for the economy and the environment [158, 159]. Using Euphorbia condylocarpa [160] and M. bieb root extract as reducing agents and stabilisers, Mahmoud Nasrollahzade and colleagues develop green synthesis of Pd/Fe3O4 nanoparticles and their catalytic uses in ligand- and copper-free Sonogashira. This method's benefits include excellent yields, a straightforward process, and ease of setup. A magnet may be used to recover the catalyst, and it can then be used repeatedly without significantly losing its catalytic activity.

According to Miran Kim and colleagues, Pd nanoparticles were successfully immobilised on a newly created core-shell Fe3O4@aminefunctionalized graphene (Fe3O4@GON) [161]composite. Amination of GO produced the core-shell Fe3O4@GON composite. With amine groups, the oxygen functional groups of GO may be switched out with ease, and they exhibit strong cohesiveness with Pd. On Fe3O4@GON, monodisperse Pd nanoparticles were created after the Pd nanoparticles were immobilised through sonochemical reduction. In the Sonogashira cross-coupling reaction between aryl iodides and terminal acetylenes, the produced Pd/Fe3O4@GON composite served as the catalyst.

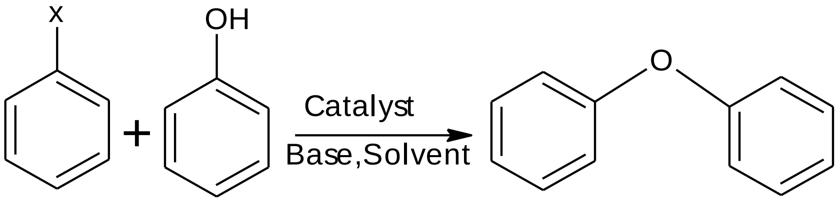
Materials science and engineering have recently paid a lot of attention to graphene quantum nanodots (GQD), which have special properties such as low toxicity, excellent photostability, small size, biocompatibility, highly tunable photoluminescence properties, chemical inertness, exceptional multi-photon excitation property, electrochemiluminescence, and ease of functionalization with biomolecules. The synthesis of PdCu [162] bimetallic NPs supported by GQD modified Fe3O4 NPs and their use as an effective recyclable catalyst for the Sonogashira reaction of aryl iodides, bromides, and chlorides was reported by Mohammad Gholinejad and coworkers.

For the preparation of tetrazole-functionalized nanomagnetic Fe3O4-supported palladium nano-particles and their use in the copper and phosphine-free Sonogashira reaction, Ali Elhampour and colleagues provide a successful and broadly applicable methodology[163]. The developed catalyst exhibits outstanding Sonogashira reaction catalytic activity, high yield, and reusability.

The present study by Mohsen Esmaeilpour and colleagues describes the formation of an N-heterocyclic carbene-Pd(II) complex based on theophylline and supported by Fe3O4@SiO2[164] nanoparticles, as well as its use in environmentally friendly Sonogashira-Hagihara cross coupling processes. The significant advantages of this approach include facile product separations and purifications, fast reaction times, great yields, clear reaction profiles, low catalyst loading, effective magnetic separation, and good chemical stability of the catalyst. Up to eight cycles, the catalyst exhibits good catalytic activity.

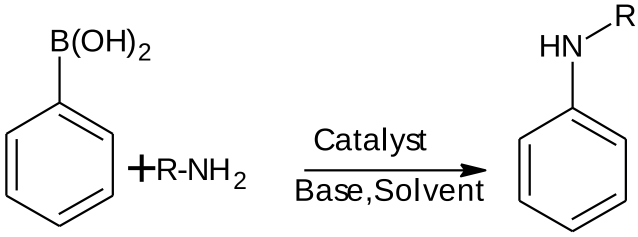
VIII. C-O COUPLING REACTION

The enormous number of natural compounds that include ether, ketone, or ester functionality and the presence of C-O bonds have sparked interest in the field of synthetic organic transformations [165]. To catalyze these processes, magnetically reusable catalysts with transition metal complexes immobilized on the surface of the magnetite core have been created[166]. The Fe3O4@SiO2@SePh@Pd(0)[167] nanocatalyst designed by Alpesh K. Sharma and colleagues exhibits high recyclability and product yield in a short amount of time.



IX. C-N COUPLING

The synthesis of a wide range of compounds, including medicines and agrochemicals, has largely relied on the C-N cross coupling reactions[168]. Fe@Pd nanowires are created by Mahmoud Nasrollahzadeh and colleagues, and their catalytic activity in the production of ligand-free C-N bonds in water is studied. The main advantage of the current approach is the direct creation of Fe nanowires from iron electrode discharge in water. The produced catalystexhibits excellent reusability and the highest possible product yield.



Applying Fe3O4 as magnetic nanoparticles (MNPs) to supported catalytic systems is a highly effective way for easily removing chitosan with an external magnet. Chitosan is a by-product of the fishing industry with free amino and alcoholic groups that may be functionalized with organic molecules [169]. Therefore, using MNPs-chitosan to coat the magnetic core effectively is a smart move. Pd/Cu Free is designed by Abdol Reza Hajipour and his coworkers[170]. Using two modified magnetic chitosan cobalt catalysts, Heck and C-N coupling reactions.

X. MULTICOMPONENT REACTIONS

Multicomponent Reactions (MCRs) are one-step processes that combine at least three reactants to produce a final product that, ideally, includes all of the initial reactants in their entirety. MCRs are a great alternative approach since they have several advantages over traditional methods. MCRs have a number of benefits, including atomic economy, reduced solvent usage, low cost, low energy consumption, and low time use. As a result, using these kinds of reactions as a method can help develop green and sustainable procedures [171]. Fe3O4 magnetic nanoparticles (MNPs) were used as catalysts or the centre of magnetic catalysts in processes (172-178). Agglomeration of magnetic nanoparticles is simple and reduces the amount of accessible active sites. This issue is fixed by changing their surface's coating [179]. MNPs are coated with a variety of materials [180]. Examples include cellulose [181], silica [182, 183], triethylamine [184], and chitosan [185–187]. MNPs can be coated with Poly(ethylene oxide) (PEO), a non-toxic, chemically stable, reasonably priced polymer [188]. PEO may be employed in environmentally friendly synthesis techniques since it has several hydroxyl groups that make it hydrophilic and soluble in water.

Piperidine, phenylacetylene, and cyclohexane carbaldehyde were utilised as standard substrates by T. Zeng and colleagues in their quest for an appropriate solvent for the Fe3O4 nanoparticle catalysed A3 -coupling[189].Tetrahydrofuran was found to be the most efficient reaction medium for this three-component coupling process after they sampled a number of solvents. Tetrahydrofuran performed well in the magnetic separation of nanoparticle catalysts as well as the coupling reaction of aldehyde, alkyne, and amine in good yield.

By using a one-pot three-component condensation of isatoic anhydride, amines, and aldehydes in the presence of catalytic quantities of Fe3O4 nanoparticles [190] in water, Zhan-Hui Zhang and colleagues establish novel synthesis techniques for key chemical compounds 2,3-dihydroquinazolin-4(1H)-ones. The catalytic activity of the catalyst may be recovered and recycled without undergoing a major loss.

Designing by Manoj B. Gawande and colleagues is simple and sustainable. Without using an additive or supplementary supply of linkers, Fe3O4-cysteine MNP[191] was created. In good yields, b-amino carbonyl and hydroquinoline compounds were effectively synthesised using Fe3O4-cysteine MNPs in multicomponent processes. since of their efficacy, usability, and affordability, magnetic organocatalysts are extensively used and ecologically benign since they are easily recovered by straightforward magnetic decantation and maintain their catalytic activity after nine cycles.

Alaa M. Munshi and colleagues describe a catalyst system that uses a superparamagnetic Fe3O4 nanoparticle core that has been coated with gold (Fe3O4@Au)[192] to catalyse the production of propargylamine through the A3 -coupling process. They show that this catalyst is highly recyclable and that utilising an external magnetic field, it is simple to separate from the reaction mixture.

Ali Elhampour and colleagues describe the synthesis of new, functional, and recyclable Ag nanoparticles supported on magnetic hollow-Fe3O4 cores with a mesoporous TiO2[193] shell to effectively catalyse the A3 coupling reaction for the production of propargylamine derivatives in mild reaction conditions. With the use of an external magnet, the generated highly active magnetic catalyst was simply detached and reused five times in the model process without significantly losing any of its catalytic activity.

New heterogeneous magnetic gold catalysts are created by Nasrin Zohreh and colleagues [194] based on stabilising gold nanoparticles on NNN pincer ligands generated from triazine core [195,196]. For the synthesis of propargyl amines, they specifically studied the pincer-Au catalysed one-pot reaction of terminal alkynes, secondary amines, and aldehydes. The highly active magnetic catalyst was created, readily separated using an external magnet, and reused for several reaction cycles.

New heterogeneous catalysis of A3 coupling processes is designed by M. Gholinejad and colleagues. They discovered that the usage of novel magnetic NPs modified with a pyridyl-triazole ligand might help stabilise gold NPs and be used as an effective heterogeneous[197] recyclable catalyst for the synthesis of propargylamine through an A3 coupling process.

In order to create a novel, reusable, and magnetically recoverable catalyst, Zeinab Zarei and colleagues attached Zn(II) onto the magnetic natural hydroxyapatite (ZnII/HAP/Fe3O4)[198]. In a one-pot, three-component A3 -coupling reaction involving terminal alkynes, aldehyde, and secondary amines under solvent-free conditions, it has been discovered that the novel nanocatalysts can efficiently catalyse the production of structurally distinct propargylamine derivatives. The outstanding product yield, wide range of substrates, moderate reaction temperatures, reduction of chemical waste, straightforward work-up process, straightforward catalyst manufacture, and environmental friendliness are only a few of the benefits offered by this green protocol. Importantly, the produced nanocatalyst may be retrieved by using an outside magnetic field and re-used for seven times without suffering significantly from activity loss.

Fe3O4@SiO_2-$ZrCl_2$-MNPs[199], a new nano magnetic reagent, was produced by Fatemeh Kamali and colleagues. After preparation, the catalyst's ability to assist in the synthesis of 2H-indazolo[2,1-b]phthalazine-triones and tetrahydrobenzimidazo[2,1-b]quinazolin-1(2H)-ones is investigated. High yields, quick and simple catalyst preparation, moderate reaction conditions, quick reaction times, and catalyst that can be recycled for a minimum of four runs constitute all ositives.

Molybdenum Schiff base complex supported on magnetite nanoparticles is designed by Jamshid Rakhtshah and colleagues as a heterogeneous catalyst[200] for the one-pot multicomponent synthesis of -aminonitrile derivatives. This procedure is advantageous from an environmental, practical, and financial standpoint since it has a wide range of substrates, is simple to recover the magnetite catalyst from, operates in solvent-free conditions, has little reaction time, and produces excellent product yields..

For the very effective multicomponent synthesis of pyran derivatives, Ali Maleki and colleagues developed a poly(ethyleneoxide)-based magnetic nanocomposite catalyst (201).The catalyst demonstrates effective recycling in a timely manner with improved yield.

As pyrano[2,3d] pyrimidine derivatives, Sami Sajjadifar and colleagues synthesize Due to their beneficial properties as anticoagulants, diuretics, spasmolytics, anticancer, and antianaphylactics, pyrimidines and their derivatives might be considered essential substances in the field of drugs and pharmaceuticals.[202] The magnetic nanocatalyst used in this study (Fe3O4@APTES@isatin-SO3H), which is new, effective, and reusable, is coated with amino propyl to modify the nanoparticles.

Since propargyl amines are significant building blocks in both natural products and the compounds that make up medicinal drugs, they are useful synthetic intermediates in organic synthesis.[203-211]With caffeine-coated magnetic nanoparticles as support, Mohammad Gholinejad and colleagues developed a new heterogeneous catalyst called Fe3O4@CaffAu) [212] that is stable and readily available. This catalyst can be recycled magnetically for at least nine consecutive runs without suffering a significant loss of activity and with only a slight aggregation of Au.

A new cobalt Schiff-base complex immobilized on silica-coated Fe3O4 nanoparticles[213] is used as a reusable catalyst in the efficient and straightforward one-pot three-component synthesis of pyranopyrazole derivatives designed by Behrouz Shaabani and colleagues.

A. Maleki and colleagues develop a straightforward one-pot method for the multicomponent synthesis of pyrano [2,3-d]pyrimidine derivatives, which is carried out by newly created Fe3O4@polyvinyl alcohol magnetic nanoparticles[214].

Using a novel hydrothermal method, Sajjad Azizi and colleagues created paramagnetic dendritic fibrous nano-silica functionalized by aminopropyltriethoxysilan (Fe3O4@KCC-1-nPr-NH2) [215], which was used as a highly effective, recyclable, and heterogeneous nanocatalyst for the synthesis of a variety of tetrahydrodipyrazolopyridine.

Using a natural and eco-friendly support composed of copper oxide nanoparticles, Ali Maleki and colleagues create a new green approach for the synthesis of 1,2,3-triazoles [216]. Without suffering a considerable reduction in activity, catalyst may be easily recycled magnetically for numerous consecutive cycles. New magnetically reusable [217] nanocatalysts were developed by Mosstafa Kazemi for the biginelli synthesis of dihydropyrimidinones.

Novel Fe3O4@Schif-base-Cu catalyst was developed by Muhammad Aqeel Ashraf and colleagues [218] successfully. This catalyst may be used to create polyhydroquinolines in water, a green solvent. This catalyst was easily recoverable and used several times without significantly losing effectiveness..

Different magnetically separable catalyst used in various types of reaction are listed in table

Sr. No	Catalyst	Mol %	Yield %	No. of recovery cycles
Suzuki Coupling reaction				
1	SiO_2@Fe3O4@Pd	0.5	90-99	6
2	Fe3O4@SiO_2-Oxime-Pd(II)	0.3	88-91	6
3	Fe3O4@SiO_2-immine - Pd(II)	0.2	88-90	16
4	Fe3O4@SiO_2-Im-Phos-Pd	0.3	88-90	6
5	Fe3O4@CS-SB-Pd	0.2	90-99	8
6	Fe3O4-IL-Pd	0.1	88-95	10
7	Fe3O4@SiO_2-mSiO_2-Pd(II)		90-99	6
8	Fe3O4- rGO-Pd	0.2	90-95	8
9	Fe3O4 DA-Pd(II)/Pd(0)	0.2	88-95	5
10	Fe3O4@SiO_2-DTZ-Pd	0.5	90-95	5
11	Fe3O4-Ethyl -CN-Pd	0.3	90-96	8
12	Fe3O4@SBTU-Pd	0.5	80-90	5
13	Fe3O4@ABA-Pd(0)	0.56	80-98	6
14	Fe3O4@[$(EtO)_3$Si-LH]-pd(II)	0.2	90-98	12
15	Fe3O4@bigua-CS-Pd(0)/(II)	0.2	90-96	8
16	Fe3O4@SiO_2@PDA-Pd	0.2	90-98	12
17	Fe3O4@GO-Pd	0.15	90-95	6
18	Fe3O4@SiO_2-Isoniazide-Pd	0.2	65-90	8
19	Fe3O4@SiO_2-Pd	0.3	90-96	8
20	Fe3O4@rGO-Pd	0.12	90-99	6
21	Fe3O4@SiO_2-NH_2-TC-Pd	1.5	90-99	9
22	Fe3O4@GO-PAMPS-Pd	0.2	90-96	9
23	Fe3O4@Sporopollen-Pd	0.1	90-95	10
24	Fe3O4@SiO_2@3-Glycidoxypropyltrimethoxysilanetriazole@Pd	0.12	75-90	10

25	Fe3O4@SiO_2-$(CH_2)_3$-N-CH-Ar@Pd	0.1	90-95	6
26	Fe3O4@TA-Pd	0.2	70-80	10
27	Fe3O4@SiO_2@N-amidinoglycine-Pd	0.1	90-99	8
28	Fe3O4@SiO_2@VB_1-Pd	0.1	90-95	6
29	Fe3O4@Green tea extract-Pd	0.3	90-98	9
30	Fe3O4@PCT-CMC-Pd	0.5	85-90	8

Sr. No	**Catalyst**	**Mol %**	**Yield %**	**No. of recovery cycles**
Heck Coupling Reaction				
1	Fe3O4@NHC-Pd	0.56	80-95	5
2	Fe3O4@SiO_2-Polymer imd.-Pd	0.3	80-90	6
3	Fe3O4@DAG-Pd	0.3	90-98	6
4	Fe3O4@[$(EtO)_3$Si-LH]-pd(II)	0.2	90-98	12
5	Fe3O4@Pd(II)	0.5	95-98	6
6	Fe3O4@NHC-Pd(II)	0.1	88-98	6
7	Fe3O4@ABA-Pd(0)	0.56	80-98	6
8	Fe3O4@C-Pd	0.7	70-90	6
9	Fe3O4@SiO_2-T-Pd	1.9	70-94	6
10	Fe3O4@Pd	1.0	91-99	5

Sr. No	**Catalyst**	**Mol %**	**Yield %**	**No. of recovery cycles**
Sonogashira Coupling reaction				
1	Fe3O4	5	90-98	6
2	Fe3O4@SiO_2-SB-Pd(II)	0.5	90-93	5
3	Fe3O4@Pd	0.15	80-90	5
4	Fe3O4@SiO_2-Cu-Pd	0.3	80-90	6
5	Fe3O4/SiO_2/P(GMA-co-EGDMA)–PEI–Pd(0)	0.1	80-90	8
6	Pd@ Fe3O4@GON	0.4	90-99	6
7	Fe3O4@GQD-Cu-Pd	0.3	90-99	6
8	Fe3O4@SiO_2-T-Pd	0.2	80-90	8
9	Fe3O4@SiO_2-NHC-Pd(II)	0.43	90-99	6

Sr. No	Catalyst	Mol %	Yield %	No. of recovery cycles
Multi component Reaction				
1	Fe3O4	0.5	75-80	5
2	Fe3O4	0.15	78-80	5
3	Fe3O4-Cys	0.5	40-90	5
4	Fe3O4-Au	0.1	80-85	5
5	Zn_2/HAP/ Fe3O4	0.5	90-95	7
6	h-Fe3O4@m-TiO_2/Ag	0.01	45-96	5
7	MNP@Au/NNN-Pincer	0.07	90-95	6
8	Fe3O4@SiO_2-$ZrCl_2$	0.25	88-95	5
9	Fe3O4@Si@$MoO_2(acac)_2$	0.2	90-95	5
10	Fe3O4@PEO-SO_3H	0.1	80-85	6
11	Fe3O4AOTES@isatinSO_3H	0.2	87-90	6
12	Fe3O4@PT@Au	0.01	85-90	10
13	Fe3O4@Caff-Au	0.07	90-95	10
14	Fe3O4@SiO_2@CosB	0.08	95-98	8
15	PVA@ Fe3O4-Cu	0.35	80-98	6
16	Fe3O4@kcc-1-npr-NH_2	0.1	95-98	10
17	Cu_2O/Agar/ Fe3O4Cu_2O@Agar@ FE3O4	0.25	80-90	5
18	Fe3O4-MWCNT	0.3	88-98	
19	Fe3O4Schiffbase-Cu	0.25	86-100	11

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