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Silicon Nanoparticles and their Uses in Synthetic Organic Reactions.

Tamer K Khatab*, Khairy A M El-Bayouki, Wahid M Basyouni, and Wael M Tohamy.

Organometallic and Organometalloid Chemistry Department, National Research Centre, 33 El-Behouth St., Dokki, 12622, Cairo, Egypt.

ABSTRACT

The mixture of synthetic and physical changes conceivable with silica builds its adaptability and its biocompatibility makes it a moderately considerate material. Silica NPs are likewise encouraging contender for enhanced natural synthetic responses too sedate conveyance frameworks. The novel heterogeneous catalysts in light of silica backings, principally on the grounds that silica shows a few invaluable properties, for example, magnificent stability (chemical and thermal), recyclability, great availability and porosity. Thus here-in different examination articles were accounted for managing wide mixtures of natural responses or organometallic particles fused into silica NPs utilizing the Stöber or one-pot multi-component, Mizoroki-Heck, Suzuki-Miyaura, Sonogashira cross coupling, Biginelli, Hantzsch, Mannich and many other different reactions

Keywords: silica nano-particles; heterogeneous; catalysts; One-pot synthesis; Multicomponent.

**Corresponding author*

INTRODUCTION

Silicon is one of the most abundant chemical elements found on the Earth. Due to its unique chemical and physical properties, silicon based materials and their oxides (e.g. silica), over 90% of the Earth's crust is composed of silicate minerals. Nanotechnology in the most recent years has become hugely which empower researchers to work matter at the nanoscale (one thousand millionth of a meter). In this size-range, materials can show new and bizarre properties, for example, distinctive synthetic reactivity, optical or attractive properties.

Silicon nanoparticles (SiNPs) with sizes littler than (~ 5 nm) concurring the Bohr energizing range quantum dabs (QDs) [1-3] can be made by a differing qualities of courses, which may be ordered as "substance" or "physical". a) Physical courses regularly incorporate high-temperature and/or vacuum affidavit procedures and are favored when the item is to create little amounts of material for physical or electronic applications. b) Chemical highways tend to deliver material of less all around characterized sythesis and size (with a few exemptions), however create a lot of material and may be good with the conjugation of organic atoms at the molecule surface [4-8].

Silicon nanoparticles have a lot of applications which used in manufacturing of semiconductors, solar cells, photovoltaic (Li-ion battery) biomedical engineering/medicine and heterogeneous nanocatalysis [9-18]. In recent years there are many forms of silica nanoparticles can be achieved.

Silica bolstered ionic liquid (ILs) have connected as heterogeneous catalysts, chromatographic material, gadgets and ""layout"" for the arrangement of nanomaterial. These silica-IL half breeds join the advantages of ILs with the idle advantage of strong silica as sample imidazolium ILs have demonstrated to affect the arrangement of exceptionally composed nanostructures, vast particular surface ranges, high porosities and reasonable pore sizes with a thin pore size circulation [19].

Silica gel has been accounted for to be a great medium for a few invaluable natural changes under natural dissolvable free conditions, similar to the Friedel-Crafts-sort nitration of arenes and one-pot Wittig-type olefination of aldehydes [20].

Mesoporous organic-inorganic mixture materials (MSNs), an alternate class of materials ordered by expansive particular surface territories and pore sizes somewhere around 2 and 15 nm, have been found by the coupling of inorganic and natural segments by layout combination. The mix of functionalities can be accomplished in three ways: a) Subsequent connection of natural segments onto an unadulterated silica lattice (joining), b) Synchronized response of condensable inorganic silica species and silylated natural mixes (co-buildup, one-pot blend), c) The utilization of bis-silylated natural antecedents that prompt intermittent mesoporous organosilicas (PMOs) [21, 22].

Attractive nano-impetuses, natural engineered responses have acknowledged the importance and criticalness of attractively recyclable nano-impetuses (MRNCs) in the improvement of clean strategies amid the most recent years. Attractive nano-impetuses have been concentrated on in different noteworthy conventions in natural science on the grounds that they are modest, strong, and can be effectively arranged regularly from base metal (iron oxide) and most altogether can be reused for a few keeps running with no loss of selectivity and action of impetuses [23a].

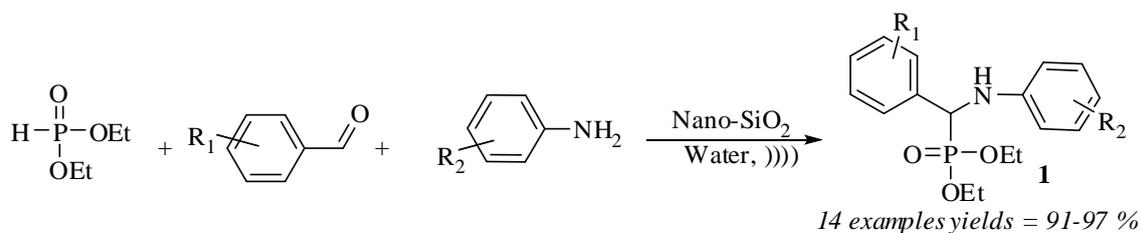
The heterogeneous catalysts in light of silica backings, essentially on the grounds that silica shows some significant properties, for example, astounding dependability (concoction and warm), recyclability, great openness and porosity. Likewise natural gatherings can be firmly connected to the surface to give reactant focuses [23b].

Along these lines and our research group interest [24], in this audit article a trial was made to survey silica nanoparticles and their applications in natural responses. A large portion of the reported information concerning the methods of combination, concoction responses also of the reported compound responses utilizing Silica-NPs amid some late year

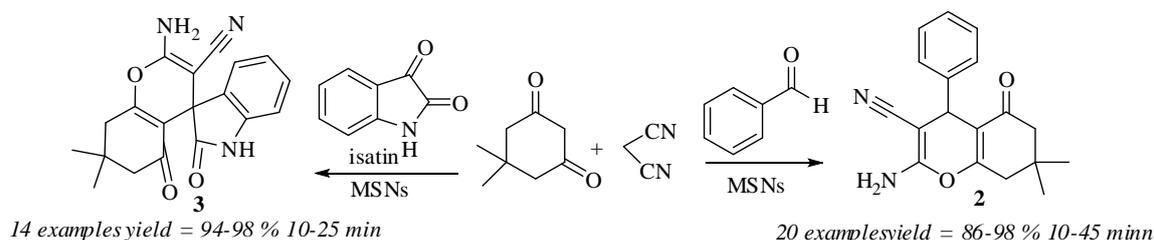
SNPS AND THEIR APPLICATION IN ORGANIC REACTIONS

Nano silica catalyst

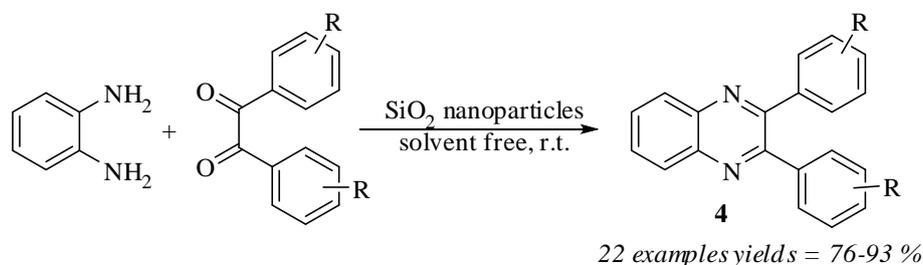
A straightforward system has been assigned for the union of α -aminophosphonates **1** in brief time and brilliant yield by the response of diethylphosphate, aldehydes and amines by means of one-pot, three-segment response utilizing nano-SiO₂ as an impetus (1.1 mol %) in H₂O as dissolvable under ultra-sonication (Scheme 1). The impetus was of high synergist movement and recoverable from the response blend utilizing filtration and reuse without noteworthy misfortunes in action furthermore eco-accommodating traits of this catalytic system [25].

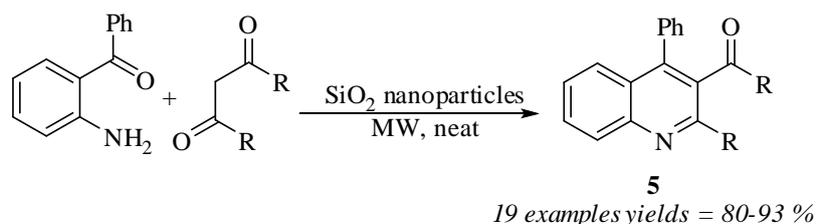

 Scheme 1: synthesis of α -aminophosphonates

Well-Ordered Mesoporous Silica Nanoparticles as a Recoverable Catalyst for One-Pot Multicomponent Synthesis of 4H-Chromene Derivatives. A simple and efficient protocol was demonstrated for the synthesis of functionalized 4H-chromenes **2**, **3** through using mesoporous silica nanoparticles (MSNs) catalyst in three component one-pot condensation reaction. The catalyst can be recycled and reused without losing activity (Scheme 2) [26].


 Scheme 2: MSNs-catalyzed synthesis of functionalized 4H-chromenes **2** and **3**

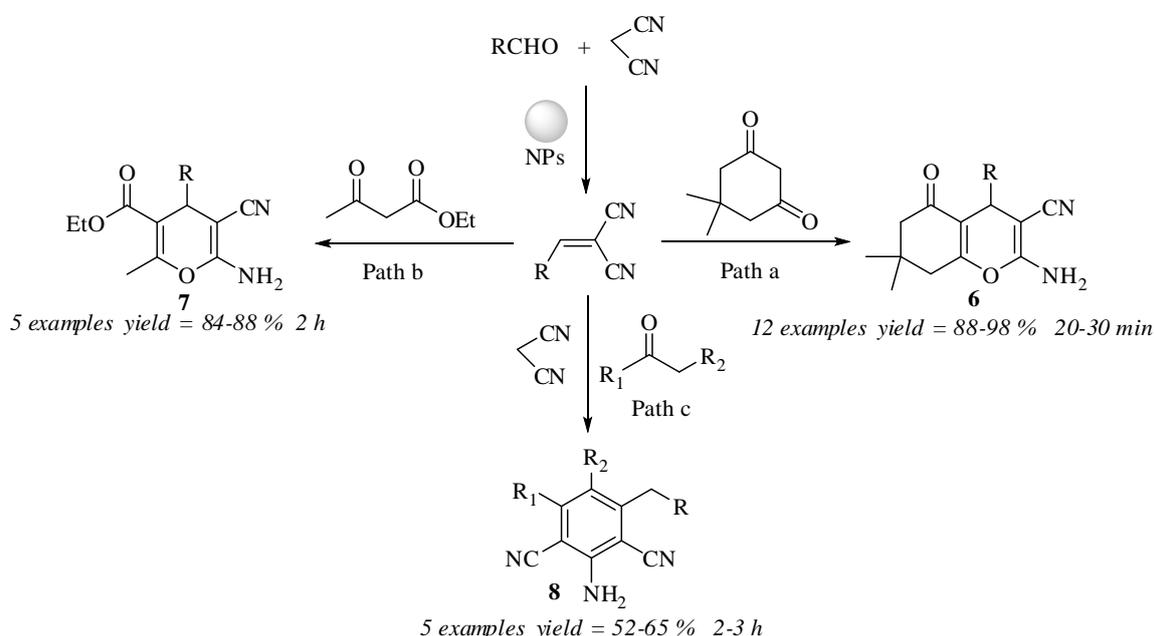
A basic system has been designed for the combination of quinoxalines and quinolines. The reaction of benzene-1,2-diamine with benzil in the presence of catalytic amount of silica (NPs) at room temperature solvent free gave high yields of quinoxalines **4** (Scheme 3). Friedlander's hetero-annulation reaction between 2-aminoaryl ketones and carbonyl compounds in the presence of silica (NPs) gave quinolines in high yields **5** under microwave MW (100-700W, 100°C) in short reaction times (Scheme 4) [27].


 Scheme 3: Synthesis of quinoxalines using SiO₂ nanoparticles at room temperature



Scheme 4: Synthesis of quinolines using SiO₂ nanoparticles under MW

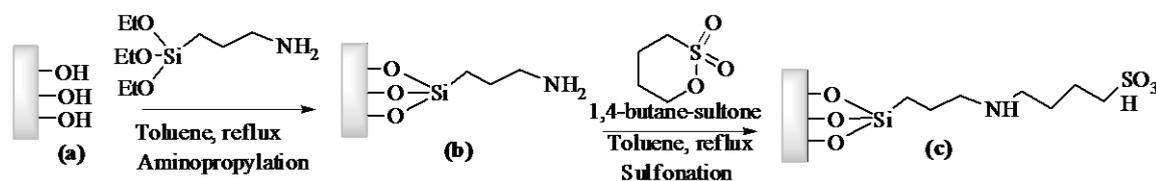
A proficient and naturally multi segment amalgamation of 4H-pyrans and poly-substituted anilines of organic, pharmacological and optical applications has been produced utilizing an exceptionally mellow, impartial and recyclable silica nanoparticles (NPs) as impetus. The 4H-pyran subordinates were blended by means of a three part response of an aldehyde, malononitrile and 5,5-dimethyl-1,3-cyclohexanedione or ethyl acetoacetate. Alternatively, poly-substituted anilines were synthesized via a four component reaction of an aldehyde, a ketone and two equivalents of malononitrile in ethanol (Scheme 5) [28].



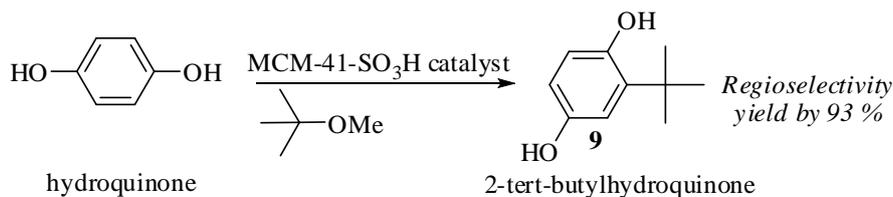
Scheme 5: SiO₂ NPs catalyzed MCRs leading to 4H-pyrans and polysubstituted anilines

SILICA IN MOBIL CRYSTALLINE MATERIAL-41

Sulfonic corrosive functionalized Mobil Crystalline Material-41 (MCM-41) as strong corrosive impetus has been performed for Friedel-Crafts tert-butylation of hydroquinone improved by microwave warming (300W). The high synergist movement of MCM-41-SO₃H was exhibited since a high hydroquinone change with a high segregation yield (93.1%) to 2-tert-butylhydroquinone (2-TBHQ) was acknowledged after 8 min. In addition, only small quantity of 2,6-di-tert-butylhydroquinone (2,6-DTBHQ, 4.0%) and other byproducts (2.9%) were noticed (Schemes 6 and 7) [29].

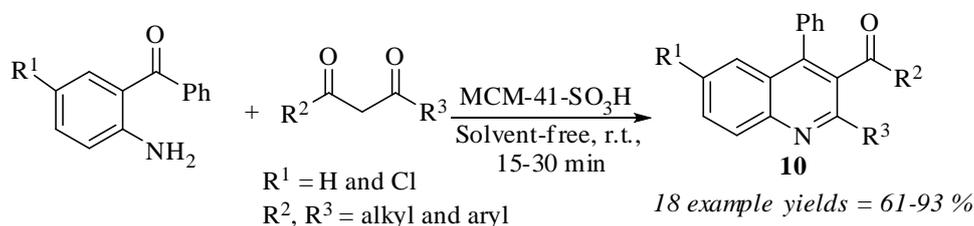


Scheme 6: The preparation of SO₃H-MCM-41 (c) catalyst from Si-MCM-41 (a)



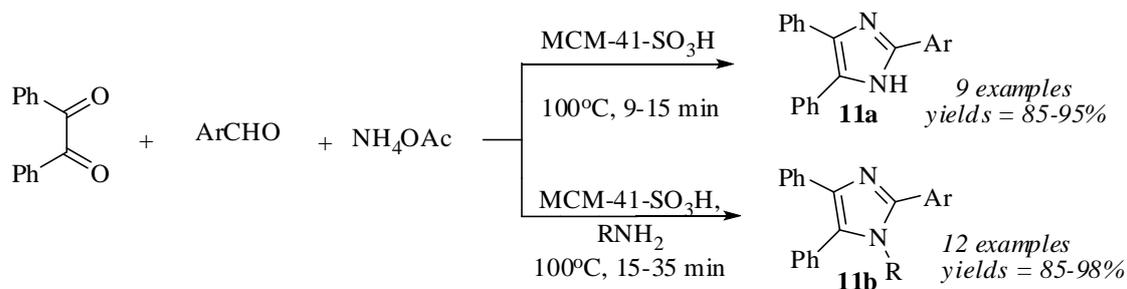
Scheme 7: Catalytic microwave-assisted Friedel-Crafts tertbutylation of hydroquinone as the probe reaction.

Quinolines **10** were synthesized via sulfonic corrosive functionalized Mobil Crystalline Material-41 (MCM-41) as strong corrosive impetus has been performed for Friedel-Crafts tert-butylation of hydroquinone improved by microwave warming (300W). The high synergist movement of MCM-41-SO₃H was exhibited since a high hydroquinone change with a high segregation yield (93.1%) to 2-tert-butylhydroquinone (2-TBHQ) was acknowledged after 8 min, at room temperature (Scheme **8**). [30].



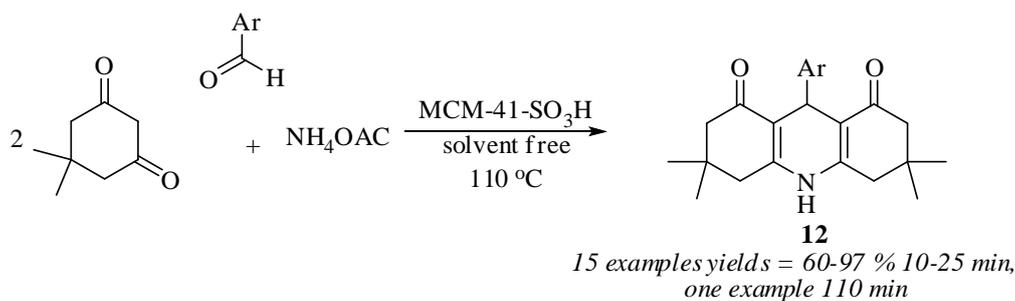
Scheme 8: Synthesis of substituted quinolones in the presence of MCM-41-SO₃H

Some imidazole derivatives can be obtained by multicomponent reaction of benzil, aldehydes and amines in the presence of metal nanoparticles as a catalyst. The Brønsted acid nanoreactor, MCM-41-SO₃H, was used in the solvent-free synthesis of trisubstituted or tetrasubstituted imidazoles **11a, b** (Scheme **9**). In this reaction, it was found out that the solvents have no role on the synthesis of the aimed imidazoles. [31].



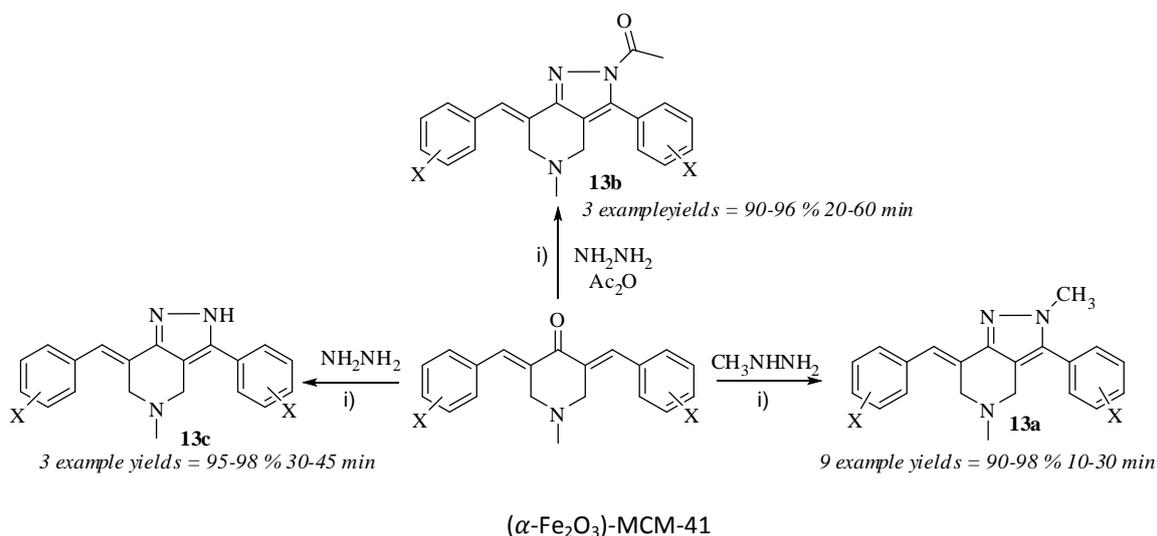
Scheme 9: Synthesis of imidazoles under solvent-free condition

MCM-41-SO₃H as a nanoreactor for the one-pot, solvent-free synthesis of 1,8-dioxo-9-aryl decahydroacridines. An efficient protocol has been established for the synthesis of 1,8-dioxo-decahydroacridines **12** from aromatic aldehyde, dimedone (2moles) and ammonium acetate in the presence of (0.005) g of MCM-41-SO₃H as a nanoreactor for the one-pot and solvent-free condition at 110°C (Scheme **10**) [32].



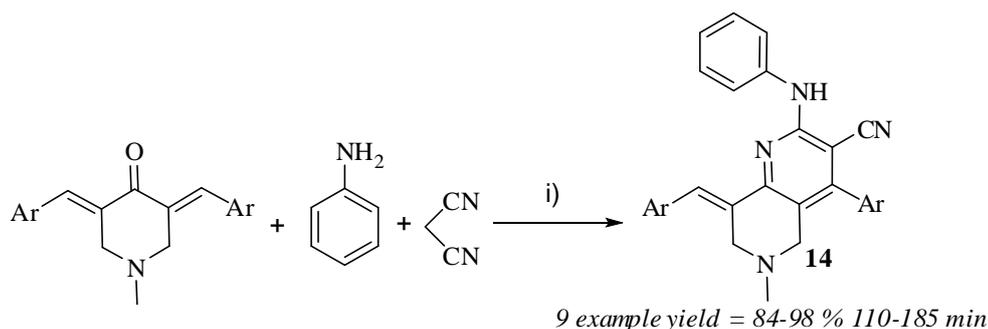
Scheme 10: Synthesis of decahydroacridines under solvent-free condition

(α -Fe₂O₃)-MCM-41 as a magnetically recoverable nanocatalyst for the synthesis of pyrazolo[4,3-*c*]pyridines at room temperature. An efficient synthesis of pyrazolo[4,3-*c*]pyridines 13a,b,c using recoverable nanocatalyst at room temperature was developed from 3,5-dibenzylidenepiperidin-4-one and hydrazine derivatives. The ideal measure of impetus was observed to be 0.015 g, and further increment in the measure of impetus has no impact on rate of the response and yield. Despite the fact that immaculate MCM-41, amino-functionalized MCM-41, and Fe₃O₄ created pleasant result, the simplicity of recoverability and reusability of (α -Fe₂O₃)-MCM-41 made it preferable catalyst for the pyrazole analogue synthesis (Scheme 11) [33].



Scheme 11: Synthesis of pyrazole analogues using dibenzylidenepiperidin-4-one

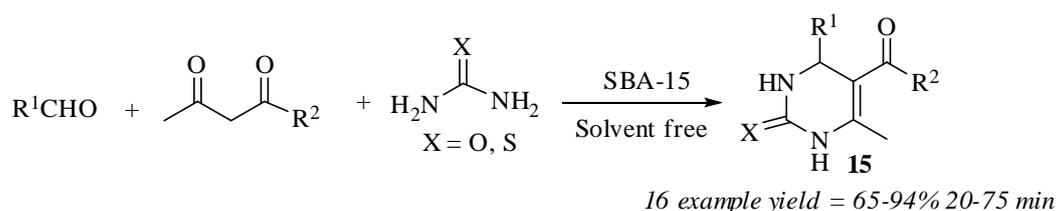
(α -Fe₂O₃)-MCM-41-SO₃H as a novel magnetic nanocatalyst for the synthesis of N-aryl-2-amino-1,6-naphthyridine derivatives. A novel magnetic (α -Fe₂O₃)-MCM-41-SO₃H acts as a nanocatalyst which could be reused even after 5 runs without decrease in activity. This acted as an efficient catalyst for the synthesis of N-aryl-2-amino-1,6-naphthyridine derivatives **14**, via reaction of aniline, malononitrile and 3,5-di(4-Cl)benzylidenepiperidin-4-one in the presence of catalytic amount of this magnetic catalyst (Scheme 12) [34].



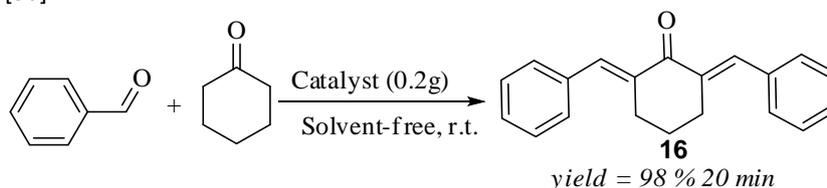
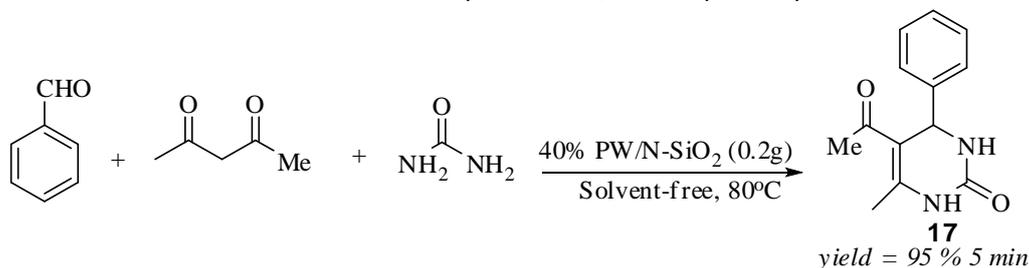
i) (α -Fe₂O₃)-MCM-41-SO₃H

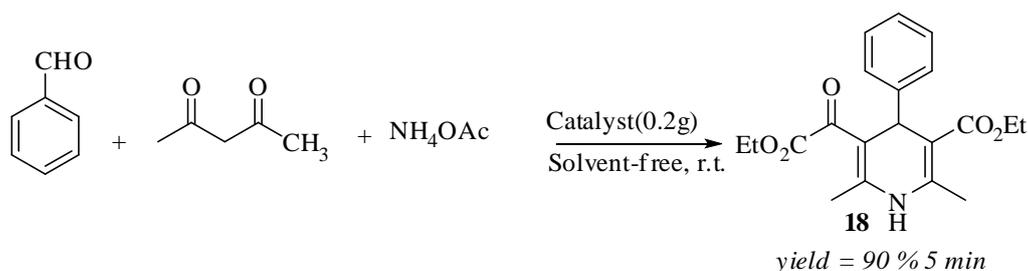
Scheme 12: Synthesis of 1,6-naphthyridine analogues
SILICA DERIVED FROM RICE HUSK ASH

A facile synthesis of 3,4-dihydropyrimidin-2(1H)-one derivatives **15** through the Biginelli reaction has been established by using heterogeneous catalyst mesoporous SBA-15 nanoparticles with pole like pore structure was blended from rice husk fiery debris (RHA) utilizing the Stem Sweep Ash (SSA) as silicon source and sol-gel technique under dissolvable free conditions. The best results were acquired utilizing 0.04 gm of mesoporous SBA-15 nanoparticles, ethyl acetoacetate, 1.5 mmol of urea and 1 mmol of aldehyde, at 100°C, under dissolvable free conditions (Scheme **13**) [35].

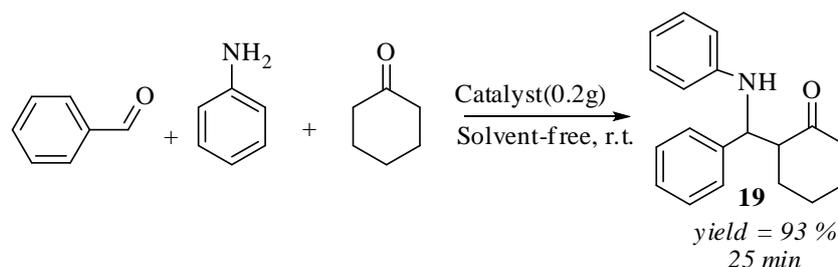

Scheme 13: Synthesis of 3,4-dihydropyrimidin-2(1H)-ones or thiones by mesoporous SBA-15 nanoparticles

An Efficient convention utilizing undefined nano-sized silica impetus was delivered from rice husk and utilized as a backing for PW (12-tungstophosphoric corrosive H₃PW₁₂O₄₀). Nano silica (PW/N-SiO₂ 40%) as a heterogeneous catalyst. PW and mass nano silica (40% PW/N-SiO₂) impetus was utilized as a part of the natural responses (Hantzsch, Biginelli, Mannich and Claisen-Schmidt responses) and displayed great action in every single synergist response, in a brief while. The impetus demonstrated higher synergist execution in correlation with PW/business SiO₂ because of its higher surface region and higher spreading of acidic protons (Schemes **14-17**) [36].


Scheme 14: Claisen-Schmidt for the synthesis of 2,6-dibenzylidenecyclohexanone 16

Scheme 15: Biginelli reaction for the synthesis of 5-acetyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one
17

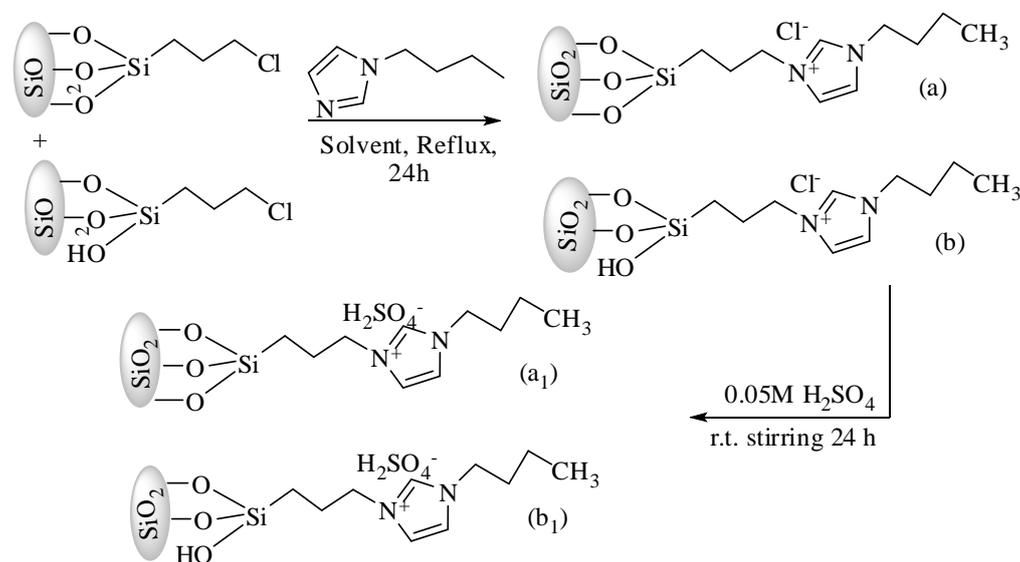


Scheme 16: Synthesis of substituted 1,4-dihydropyridine-3-carboxylate **18** via Hantzsch pyridine reaction of benzaldehyde, acetylacetone and ammoniumacetate in the presence of PW/N-SiO₂

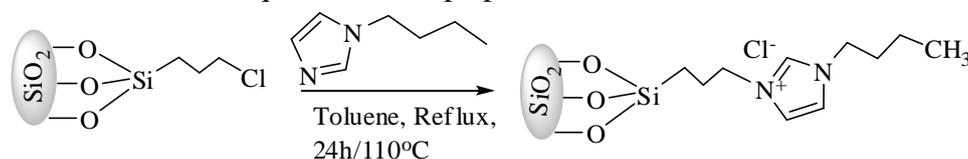


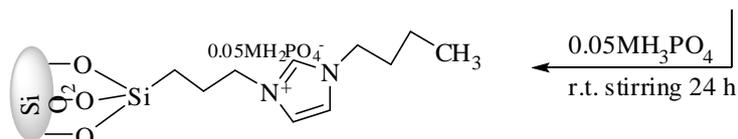
Scheme 17: Synthesis of substituted cyclohexanone **19** via Mannich reaction of benzaldehyde, aniline and cyclohexanone in the presence of PW/N-SiO₂

Rise husk cinder (RHA) was accounted for to functionalize with 3-chloropropyltriethoxy-silane (CPTES) and after that with 1-butylimidazole to make RHABIm-Chloride. The chloride particle in RHABIm-Cl was supplanted with sulfate (RHABIm-HSO₄) and phosphate (RHABIm-H₂PO₄) particle at room temperature (Schemes **18**, **19**). Catalytic performance of RHABIm-HSO₄ in acetalization of glycerol **20** with benzaldehyde give two isomers, i.e. 1,3-dioxane **21** and 1,3-dioxalane **22** (Scheme **20**). Next glycerol acetalization with a few aldehydes over RHABIm-HSO₄ demonstrated the change and selectivity of distinctive aldehydes (i.e. o-anisaldehyde, p-anisaldehyde, 2-chlorobenzaldehyde, and 2-fluorobenzaldehyde) over RHABIm-HSO₄ [37].

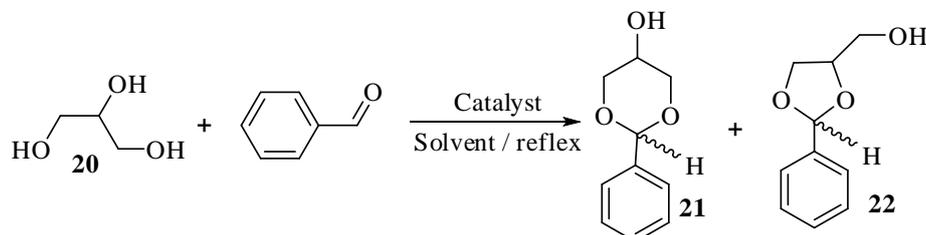


Scheme 18: the reaction sequence for the preparation of RHABIm-Cl and RHABIm-HSO₄



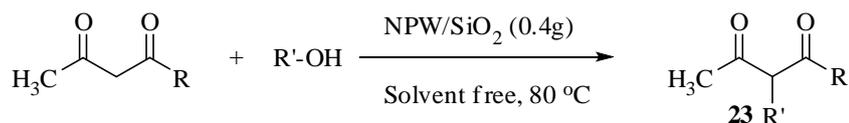


Scheme 19: the reaction sequence for the preparation of RHABIm-H₂PO₄



Scheme 20: Synthesis of the isomeric six and five member cyclic acetals of 1,3-dioxane and 1,3-dioxalane

12-Tungstophosphoric acid supported on nano silica from rice husk ash as an efficient catalyst for direct benzylation of 1,3-dicarbonyl compounds in solvent-free condition. Nano silica has been removed from rice husk, which is an agrarian waste, with high surface zone and in undefined structure. 12-Tungstophosphoric corrosive, H₃PW₁₂O₄₀ (PW) was upheld on this silica to create nano silica supported PW (NPW/SiO₂) as a nano catalyst. NPW/SiO₂ has been used as a high effective catalyst for benzylation of 1,3-dicarbonyl compounds with benzylic alcohols. The solid acid catalyst (0.4 g), was added to a mixture of alcohol (1 mmol) and 1,3-dicarbonyl compound (1 mmol) at 80 °C (Scheme 21) [38].



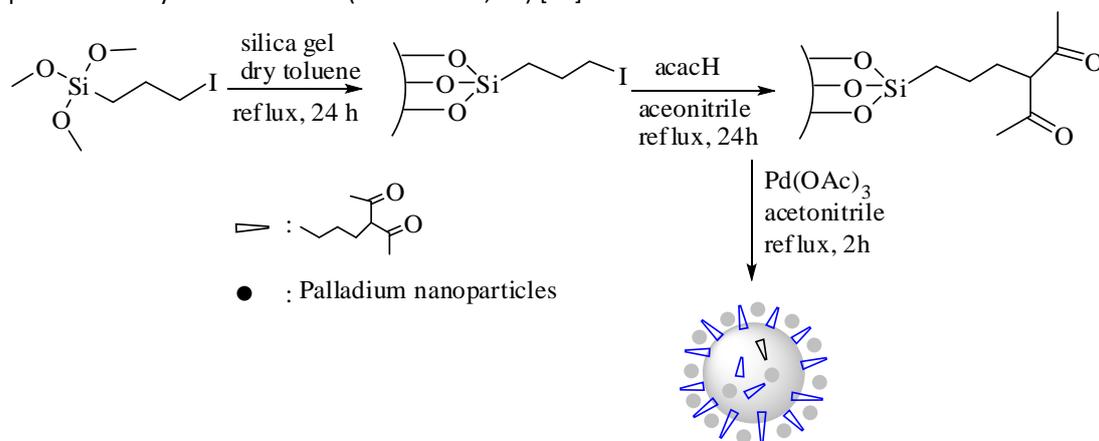
R' = benzyl derivatives

10 examples yield = 94-98 % 2-10 min

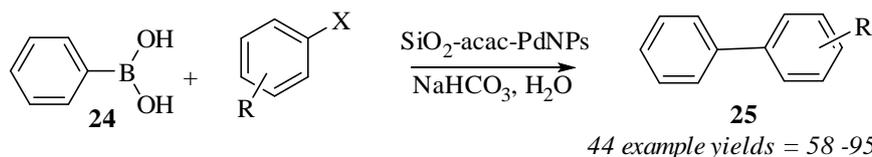
Scheme 21: Benzylation of 1,3-dicarbonyl derivatives

SILICA-SUPPORTED PD AS A CATALYST IN C-C COUPLING REACTION

Silica-acetyl (CH₃)₂CO supported palladium nanoparticles was accounted for as another heterogeneous and recyclable catalyst for Suzuki-Miyaura cross-coupling response of phenylboronic acids **24** with several aryl halides to produce the corresponding diaryl products **25** with chemo-selectivity and suitable reaction times. The response was done in H₂O as dissolvable under green conditions in the vicinity of NaHCO₃ as the base. The manufactured system for the readiness of silica-acetyl (CH₃)₂CO supported palladium nanoparticles catalysts is showed in (Schemes 22, 23) [39].

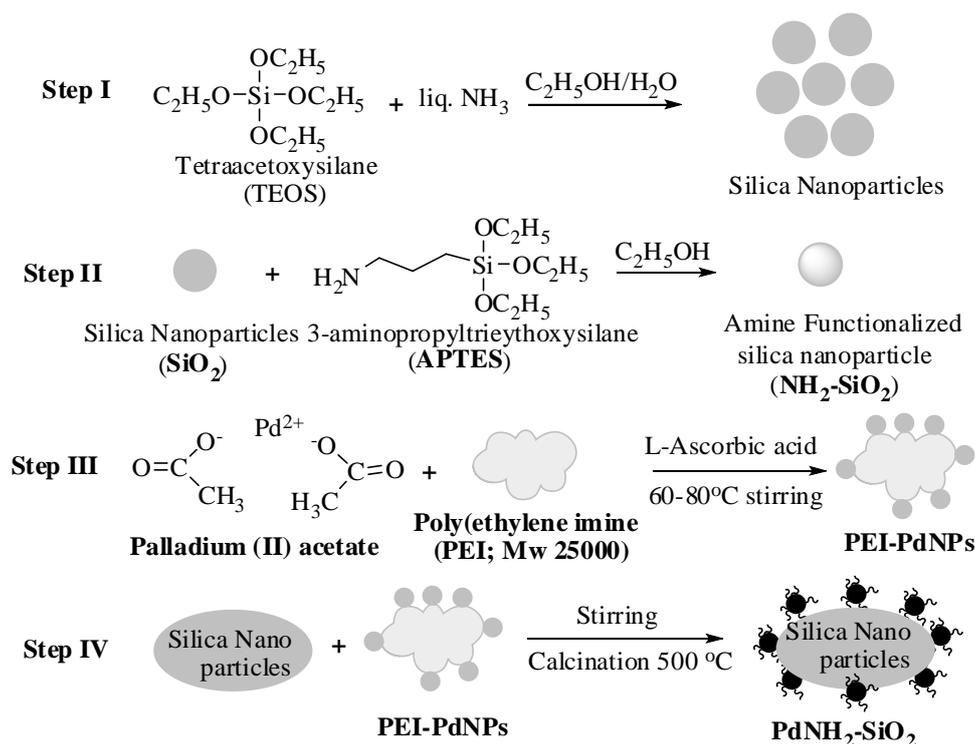


Scheme 22: synthetic route for the catalyst preparation

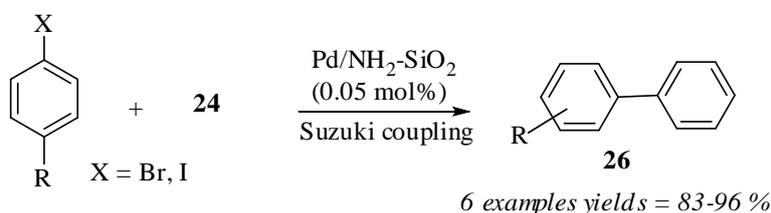


Scheme 23: Suzuki reaction of different types of aryl halides with phenylboronic acids

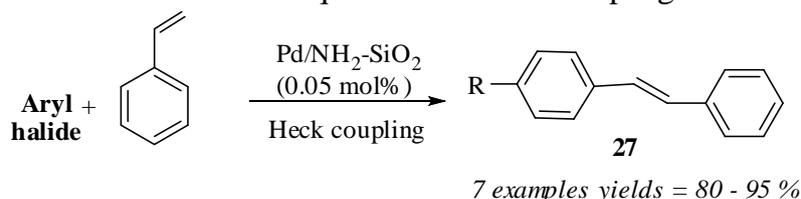
A versatile protocol was reported for the preparation of palladium nanoparticles (PdNPs) impetus immobilized on amine-functionalized silica nanoparticles (Pd/NH₂-SiO₂). The silica upheld PdNPs was orchestrated by utilizing polyethyleneimine (PEI) as a topping operators and ascorbic corrosive as decreasing specialists in fluid arrangement. This heterogeneous impetus was utilized for the Suzuki, Heck and Sonogashira coupling responses. (Schemes 24-27) [40].



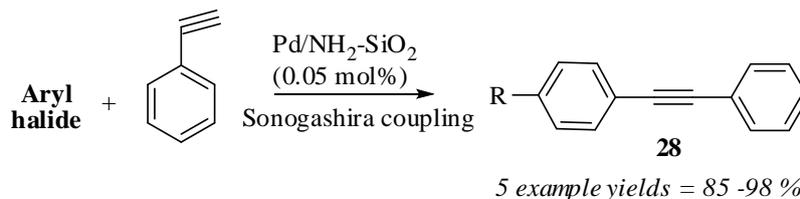
Scheme 24: Schematic representation of four-step process for the synthesis of PdNH₂-SiO₂



Scheme 25: General equation for Suzuki coupling reaction

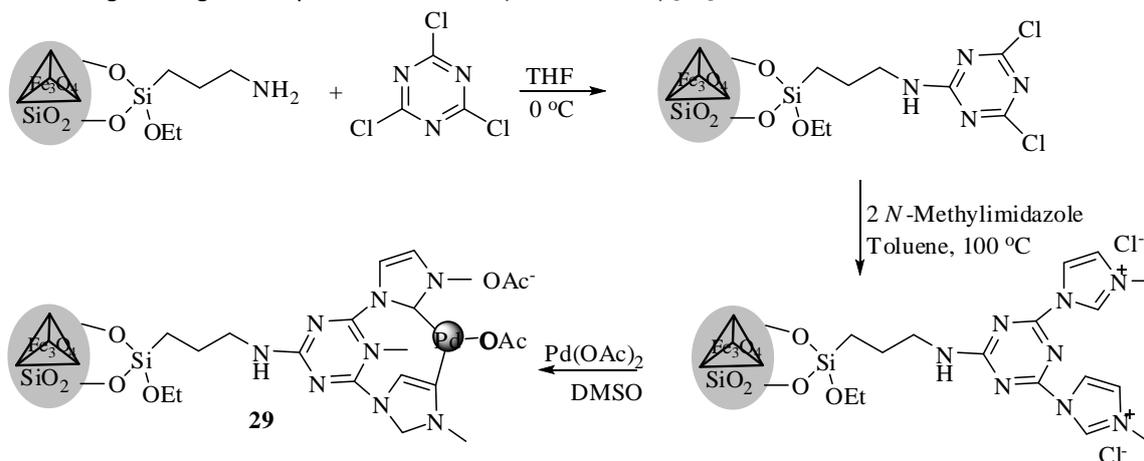


Scheme 26: General equation for Heck coupling reaction

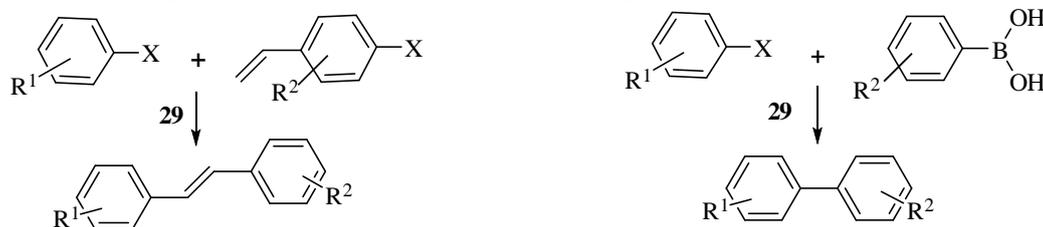


Scheme 27: General equation for Sonogashira coupling reaction

SPIONs-bis(NHC)- palladium(II) diacetate [superparamagnetic iron oxide nanoparticles] a bi-dentate NHC ligand as a capable nanocatalyst complex has been readied from silica-covered magnetite nanoparticles ($\text{SiO}_2@Fe_3O_4$) **29** (Scheme **28**) and utilized as a part of the Mizoroki–Heck and Suzuki–Miyaura coupling responses (Scheme 29). The items were delivered in exceptional returns and microwave-helped C-C coupling light at to a great degree low palladium amount (~ 0.002 mol %) [41].

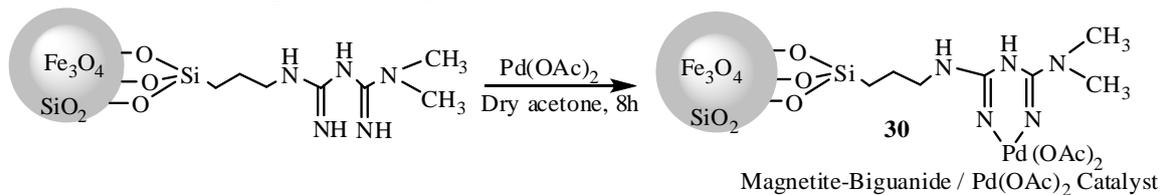


Scheme 28: preparation of SPIONs-bis (NHC)-palladium(II) diacetate catalyst

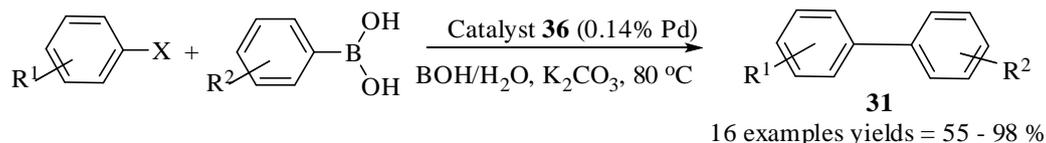


Scheme 29: Mizoroki-Heck cross coupling of aryl halides with styrene yields = 77-99 % and aryl halides with arylB(OH)₂ yields = 81-91 %

A metformin-functionalized Fe_3O_4/SiO_2 -Met-Pd(OAc)₂ as recoverable magnetite nano molecule bolstered biguanide palladium acetic acid derivation complex **30** was readied (Scheme **30**) and found as profoundly effective impetus for the Suzuki coupling response of aryl halides with arylboronic acids to yield the relating coupling items **31** (Scheme **31**). The surface-modified nanoparticle was characterized by many techniques. Simply, by using external magnet the catalyst recovered in a very short time (<15 s) [42-44].



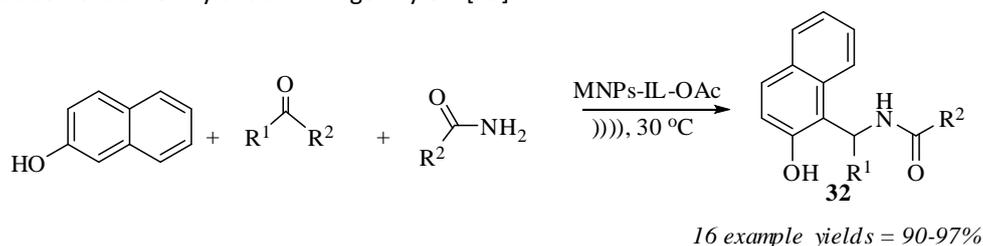
Scheme 30: preparation of Fe_3O_4/SiO_2 -Met-Pd(OAc)₂



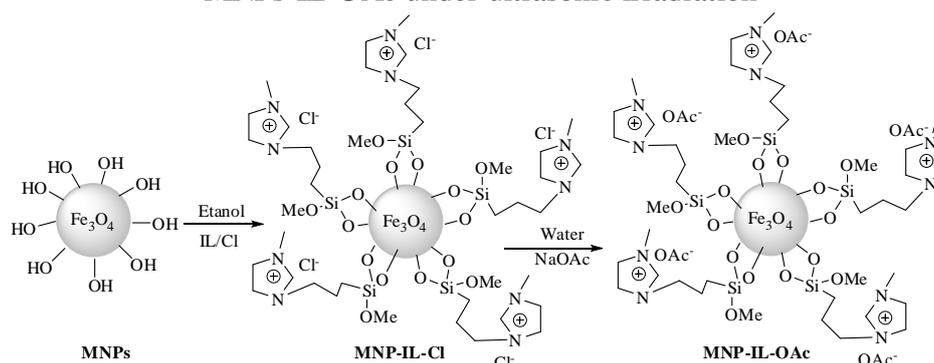
Scheme 31: Magnetic-supported biguanidine/Pd(OAc)₂ catalyzed Suzuki reaction in aqueous medium

SILICA SUPPORTED IRON OXIDES (MAGNETIC NANOPARTICALES)

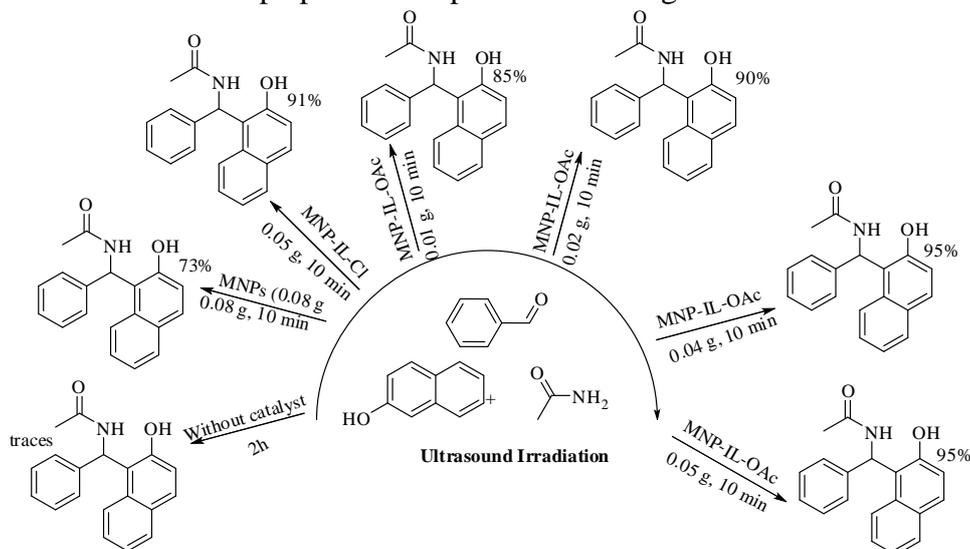
The preparation of 1-amidoalkyl-2-naphthol **32** by means of the one-pot of aldehyde, β -naphthol and amides or urea in the vicinity of attractively nanoparticles-ionic fluid acetic acid derivation (MNPs-IL-OAc, 0.04g) catalyst under ultrasound illumination (Scheme **32**). For the surface alteration, the MNPs were covered with N-methyl-3-(3-trimethoxysilylpropyl) imidazolium acetic acid derivation to create MNP-IL-OAc nanoparticles (Schemes **33**, **34**). The ideal measure of MNP-IL-OAc nanoparticles was (0.04), expanding of this sum did not demonstrate any critical change in yield [45].



Scheme 32: One-pot synthesis of 1-amidoalkyl-2-naphthol catalyzed by MNPs-IL-OAc under ultrasonic irradiation

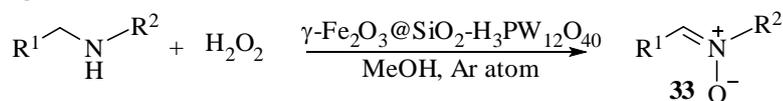


Scheme 33: preparation steps for fabricating MNPs-IL-OAc



Scheme 34: optimization for the reaction

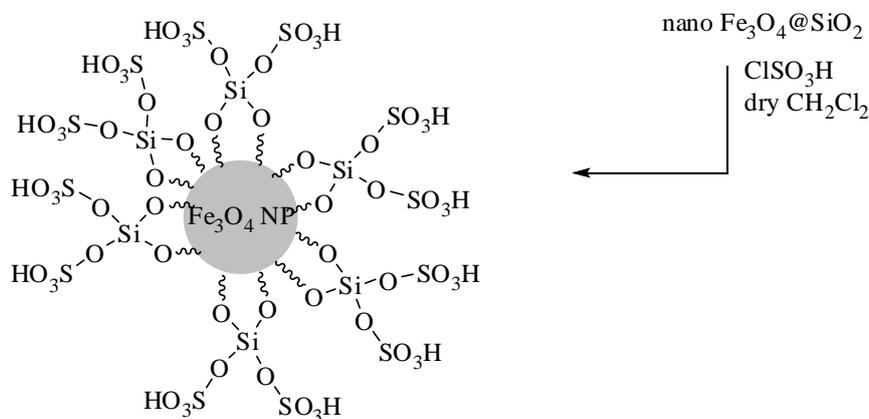
Another convention for the particular oxidation of optional amines to nitrones **33** utilizing H_2O_2 has been depicted. The super paramagnetic tungsto-phosphoric corrosive bolstered on silica-epitomized nanoparticles ($\gamma-Fe_2O_3@SiO_2-H_3PW_{12}O_{40}$) can be essentially reused. The reaction do by to oxidation steps followed by dehydration step the response completed after 1-12 h creating great yields of the nitrones (Scheme **35**) [46].



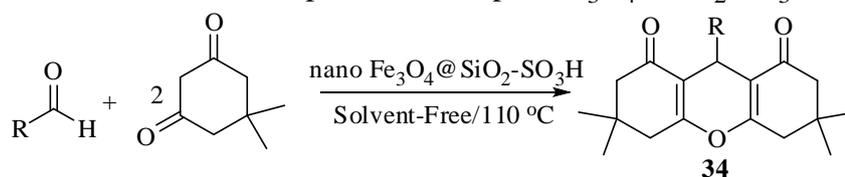
8 examples yields = 55-85 2-12 h

Scheme **35**: Synthesis of nitrones in the presence of Gama- $Fe_2O_3@SiO_2-H_3PW_{12}O_{40}$

Silanization reaction has been used to prepare Fe_3O_4 nanoparticles coated with tetraethoxysilane (TEOS). Joining of chlorosulfonic corrosive on the $Fe_3O_4@SiO_2$ nanoparticles gave sulfamic corrosive functionalized MNPs ($Fe_3O_4@SiO_2-SO_3H$) impetus (Scheme 36). Union of 1,8-dioxo-octahydroxanthenes **34** under dissolvable free conditions utilizing MNPs impetus for the one-pot (Scheme 37). Intensifies **34** were readied by means of the responding dimedone with different sweet-smelling aldehydes under dissolvable free [47, 48].



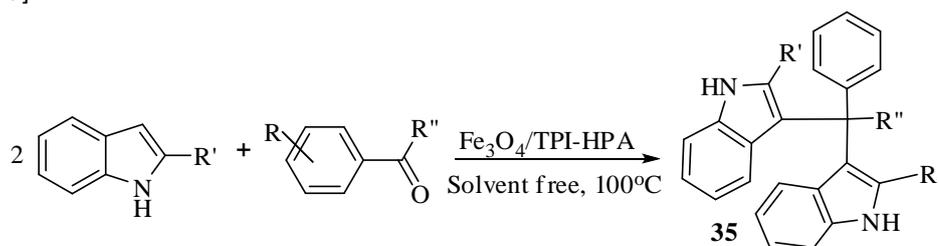
Scheme **36**: Preparation of step of $Fe_3O_4@SiO_2-SO_3H$



18 example yields 88-97 % 4-8 min

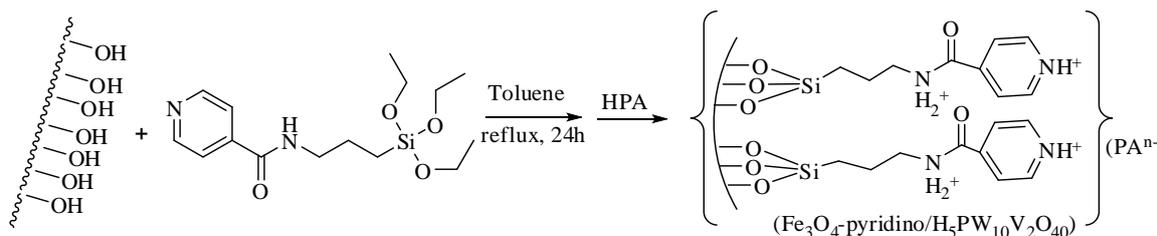
Scheme **37**: Synthesis of 1,8-dioxo-octahydroxanthenes

A new and a highly efficient catalyst $H_5PW_{10}V_2O_{40}/\text{pyridino-}Fe_3O_4$ (HPA/TPI- Fe_3O_4) nanoparticles was utilized for quick and effective amalgamation of a wide range substituted-bis(indolyl)methanes **35** by means of electrophilic substitution of indole with different ketones or aldehydes under solvent-free conditions (Schemes **38, 39**) [49].



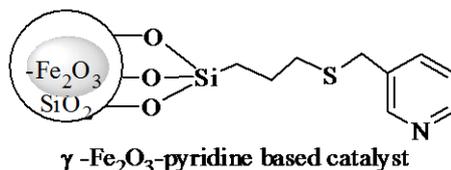
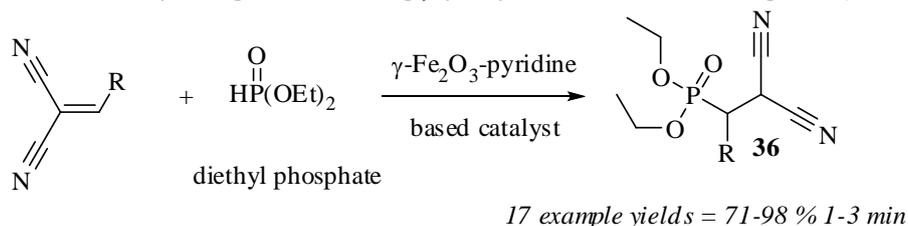
19 example yields = 45-96 % 10-40 min

Scheme **38**: General route for the preparation of different bis(indolyl)arylmethans



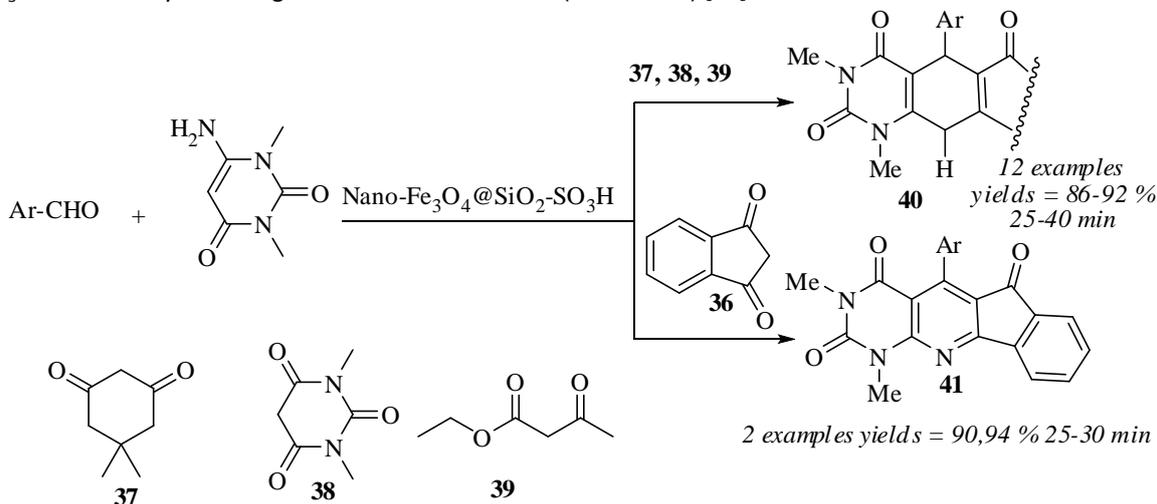
Scheme 39: Preparation of HPA/TPI-Fe₃O₄ nanocatalyst

A productive combination of β -phosphonomalonates by means of phospho-Michael expansion response of diethyl phosphate with α,β -unsaturated malonates substituted with assorted electron-pulling back and electron-giving gatherings utilizing (5 mol %) γ -Fe₂O₃-pyridine impetus under dissolvable free conditions, at 70°C. The response gave the relating β -phosphonomalonates **36** in signfica (Scheme 40) [50].



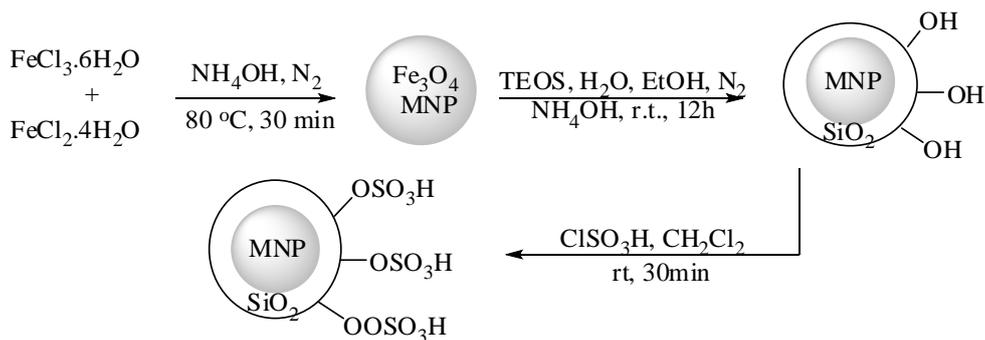
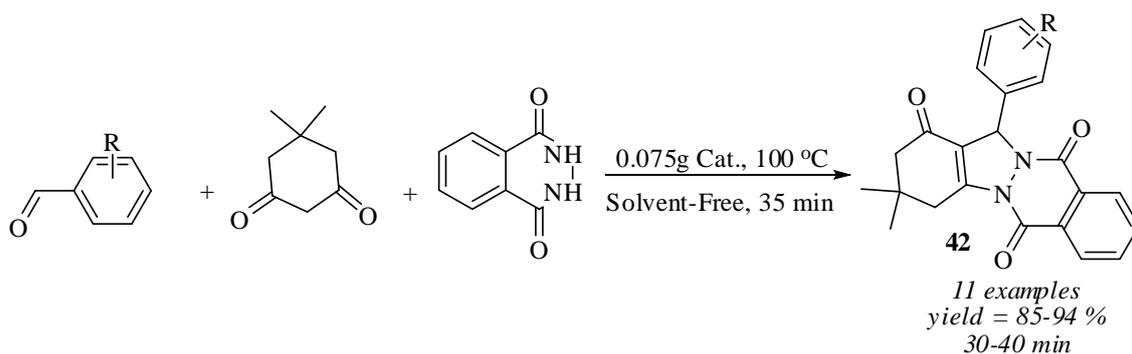
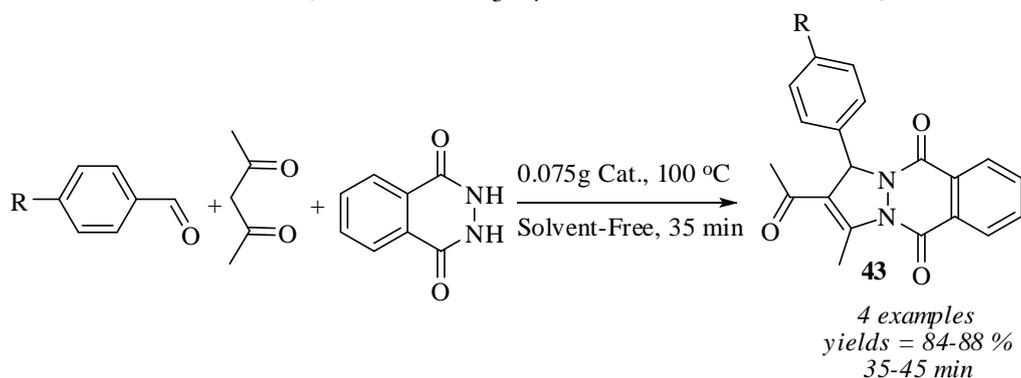
Scheme 40: Michael addition of diethyl phosphate to benzylidene malononitrile

An efficient protocol A proficient convention was created for one pot, three part blend of pyrimido[4,5-*b*]quinolines **40** and indeno combined pyrido[2,3-*d*]pyrimidines **41** utilizing nano-Fe₃O₄@SiO₂-SO₃H as the catalyst under gentle conditions in water (Scheme 41) [51].

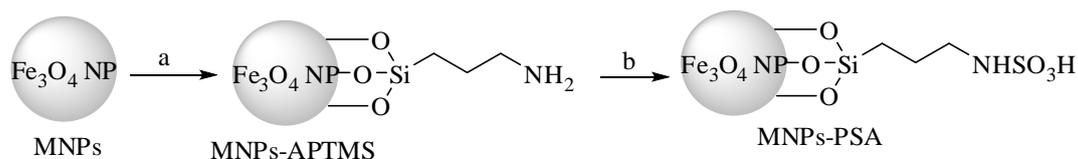


Scheme 41: Synthesis of pyrimido[4,5-*b*] and indeno fused pyrido[2,3-*d*]pyrimidines using Fe₃O₄@SiO₂-SO₃H in water

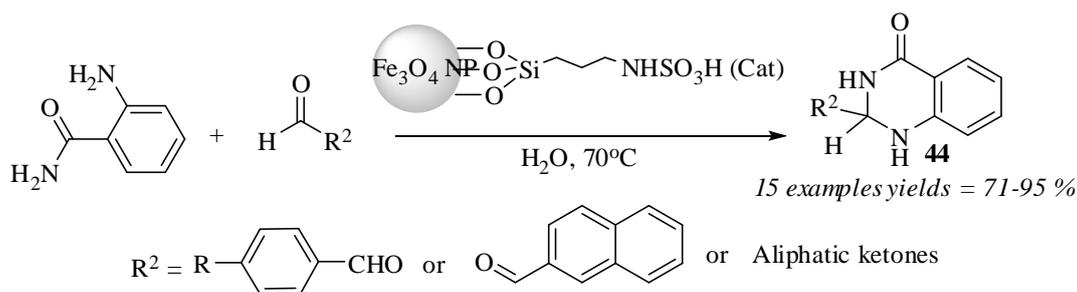
A new approach for the preparation of Fe₃O₄@silica sulfuric acid catalyst. This solid acid nano catalyst was employed for the one-pot synthesis of 2H-indazolo[2,1-*b*]phthalazine-1,6,11-triones **42** and 1H-pyrazolo[1,2-*b*]phthalazine-5,10-diones **43**. The reaction did by a three component reaction of phthalhydrazide, cyclic or acyclic diketones and aromatic aldehydes (Schemes **42**, **43**). [52].


 Preparation of Fe_3O_4 @silica sulfuric Acid

 Scheme 42: Synthesis of Fe_3O_4 @silica sulfuric acid catalyzed

 Scheme 43: Solvent free synthesis of 2*H*-indazolo[2,1-*b*]phthalazine-1,6,11-trione and 1*H*pyrazolo[1,2-*b*]phthalazine-5,10-dione catalyzed by Fe_3O_4 @silica- SO_3H

Blend of 2,3-dihydroquinazolin-4(1*H*)-ones of sort **44** has been readily through cyclocondensation of aryl aldehydes or ketones and anthranilamide in water at 70 °C in the vicinity of *N*-propyl-sulfamic corrosive upheld on attractive Fe_3O_4 nanoparticles (MNPs-PSA) as a recoverable and recyclable nano catalyst (Schemes **44** and **45**) [53].

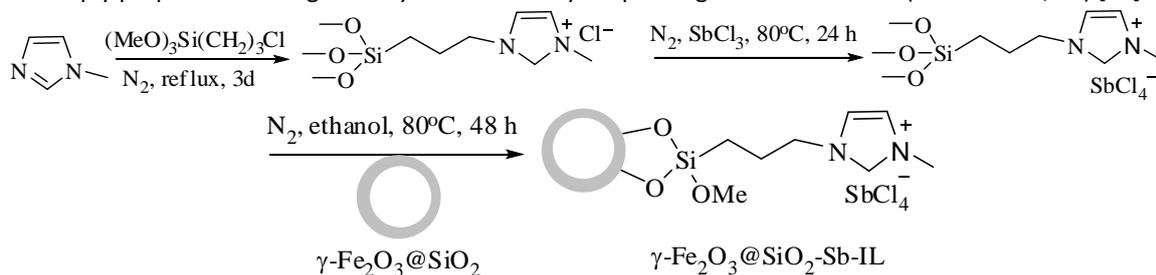


Scheme 44: Synthesis of MNPs-PSA by a) 3-aminopropyltriethoxysilane. Ethanol/water, r.t. 8 h and b) chlorosulfuric acid, dichloromethane, r.t. 2 h

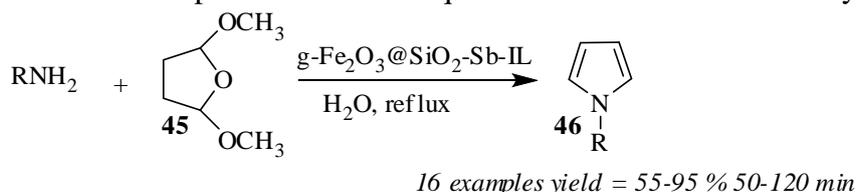


Scheme 45: MNPs-PSA catalyzes the synthesis of 2,3-dihydroquinazolin-4(1H)-ones

An efficient method has been utilized to get ready attractive nanoparticle-bolstered antimony impetus ($\gamma\text{-Fe}_2\text{O}_3\text{@SiO}_2\text{-Sb-IL}$), which displayed fantastic synergist efficiency in Clauson-Kaas response of amines to 2,5-dimethoxytetrahydrofuran **45** in aqueous medium to give N-substituted pyrroles **46**. The catalyst was simply prepared and magnetically recoverable by simple magnetic decantation (Schemes 46, 47) [54].

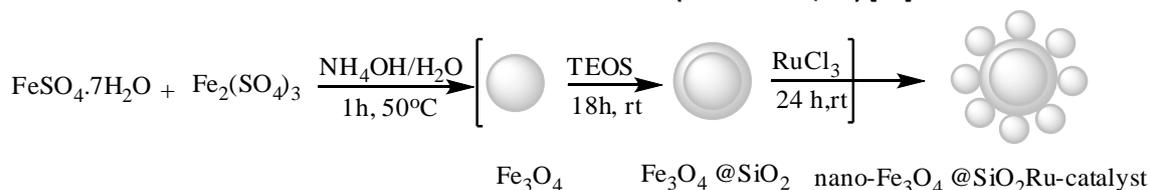


Scheme 46: Preparation of ionic liquid- $\text{Fe}_2\text{O}_3\text{@SiO}_2\text{-Sb-IL}$ catalyst

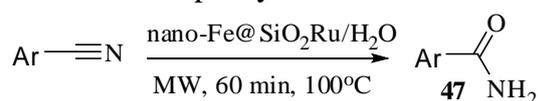


Scheme 47: synthesis of N-substituted pyrroles

Another one pot combination has been shown for the blend of attractive silica ($\text{Fe}_3\text{O}_4\text{@SiO}_2$) bolstered ruthenium (Ru) impetus; that can be promptly arranged in gram amount in one-stage under surrounding conditions in watery media (Scheme 48). The impetus was connected for the hydration of nitriles to amides **47** in fluid media under moderate conditions (Schemes 48, 49) [55].



Scheme 48. One pot synthesis of the nano- $\text{Fe}@SiO}_2\text{Ru}$ catalyst

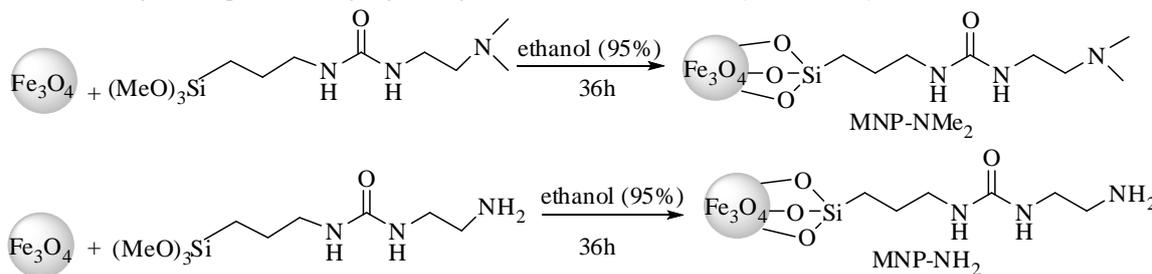


11 examples yields = 82-92 % 45 min - 2.5 h

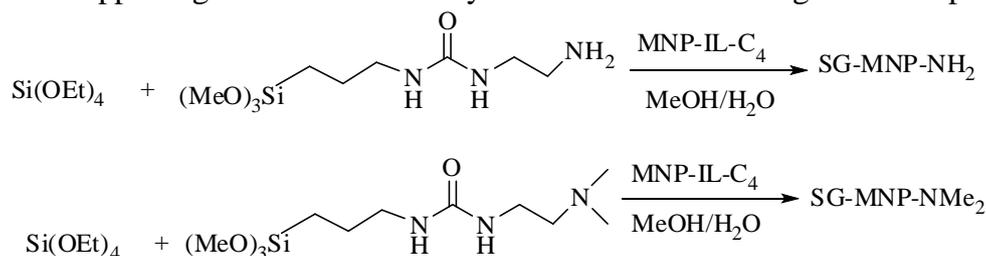
Scheme 49. Nano- $\text{Fe}@SiO}_2\text{Ru}$ catalyzed hydration of benzonitrile

Heterogeneous Catalysis vs. Quasi-Homogeneous Catalysis is Magnetically Separable Base Catalysts semi-homogeneous was orchestrated by functionalizing the surface of magnetite nanoparticles with diverse amine bunches. The Heterogeneous Catalysis was heterogeneous, arranged utilizing a sol-gel process as a part of which silane monomers functionalized with distinctive amine gatherings and poly-dense with tetraethoxysilane (TEOS) in the vicinity of magnetite nanoparticles. The first one contains primary and

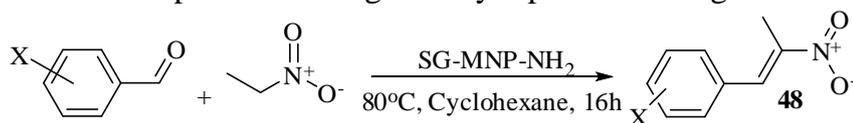
secondary amino groups (SG-MNP-NH₂) and the second one contains tertiary amino groups (SG-MNP-NMe₂) (Scheme 50, 51). The two base catalysts were used in the nitroaldol condensation of nitroethane with different aromatic aldehydes to give 2-nitroprop-1-enyl benzene derivatives **48** (Scheme 52) [56].



Scheme 50. Supporting different base catalysts on the surface of magnetite nanoparticles



Scheme 51. Preparation of magnetically separable heterogeneous base catalysts.

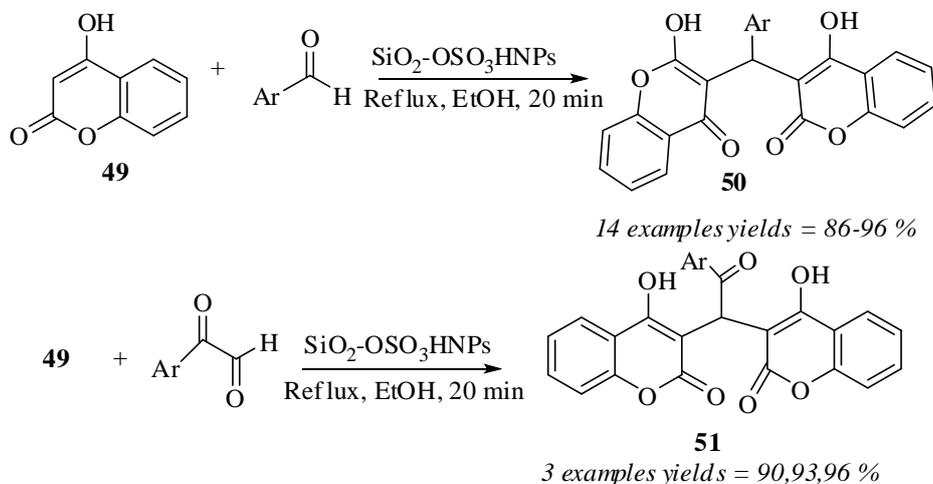


9 examples yields = 69-99 % 10-16 h

Scheme 52. Nitroaldol condensation of nitroethane with different aromatic aldehydes catalyzed by the catalyst SG-MNP-NH₂ to afford 2-nitroprop-1-enyl-benzene derivatives

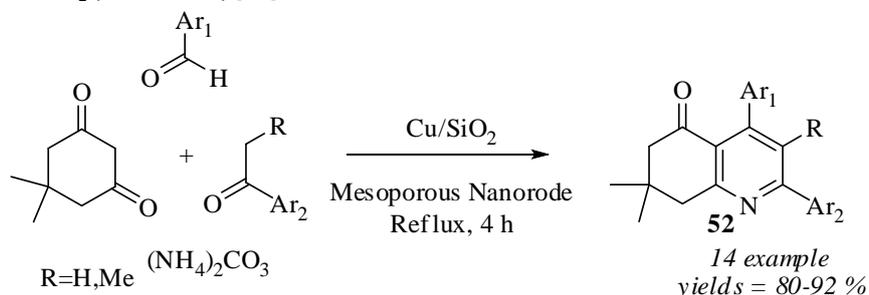
MISCELLANEOUS

A simple, efficient and green protocol has been described for the synthesis of bis-coumarins by reacting phenylglyoxals, diverse fragrant aldehydes and 4-hydroxycoumarin under reflux in EtOH in the vicinity of SiO₂-OSO₃H nanoparticles. The steady silica gel nanoparticles were essentially arranged and utilized for readiness of (SiO₂-OSO₃H NPs). (Scheme 53). [57].



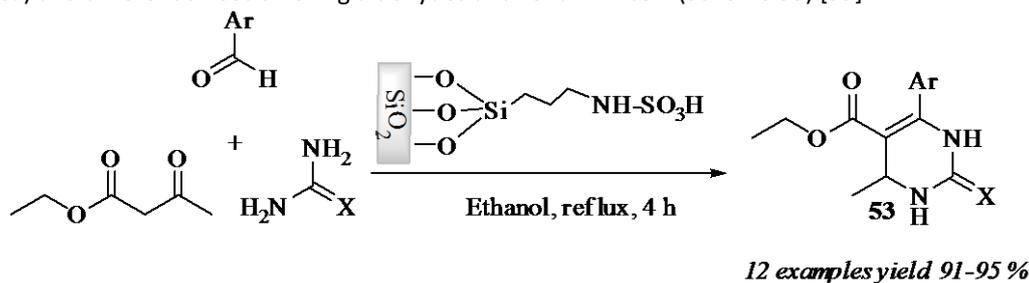
Scheme 53: Synthesis of biscoumarins using SiO₂-OSO₃H NPs as catalyst.

Copper consolidated nanorod like mesoporous silica for one pot oxygen consuming oxidative union of pyridines. One-pot, reaction of oxygen consuming oxidative union of pyridines. The responses were done in a three necked round-bottomed cup furnished with a stream of oxygen (10 ml/min). Amixture of ketone (1 mmol), 1,3-diketoneone (1 mmol), aldehyde (1 mmol) and ammonium acetic acid derivation (1 mmol) in ethanol (4 ml) was refluxed for 4 h utilizing 20 mg of the Cu/SiO₂ mesoporous nano-rode catalyst under a relentless stream of O₂ (Scheme 54) [58].



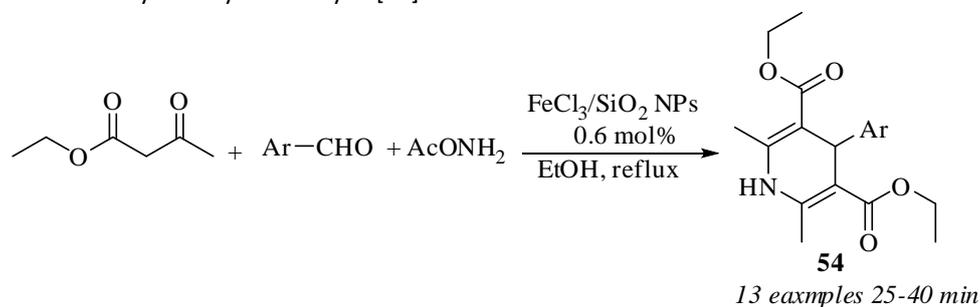
Scheme 54: One pot aerobic oxidative synthesis of pyridines

Silica-fortified N-propyl sulfamic corrosive (SBNPSA) as a heterogeneous, recyclable, strong corrosive impetus in one-pot, three parts convention for the Biginelli response. The straight forward work-up in the items' division in astounding yields, short response times, surrounding response conditions, and recyclability of bolstered impetus are amazing elements of this convention. The amalgamation of a few of 3,4-dihydropyrimidin-2(1H)- ones and - thiones **53** (91-95% yield) by means of the response of β -ketoester, urea (or thiourea) and different sweet-smelling aldehydes and reflux in EtOH (Scheme 55) [59].



Scheme 55: Synthesis of 3,4-dihydropyrimidin-2-(1H)-ones/thiones.

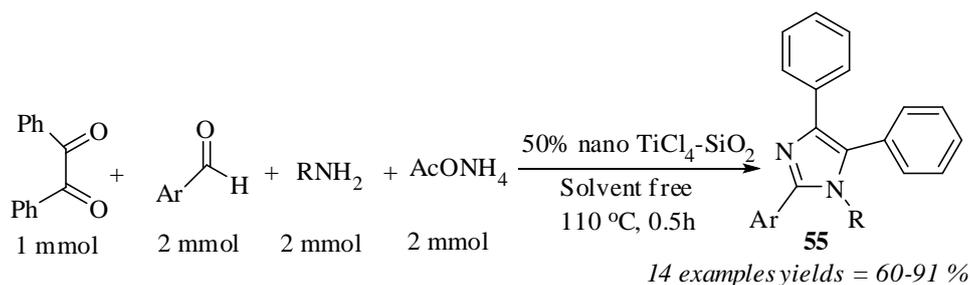
Multi-part combination of different substituted 1,4-dihydropyridines. Using silica nanoparticles (NPs) upheld Fe (III) as a shabby catalyst was executed as heterogeneous strong corrosive in (0.6 mol %) for Condensation of aldehydes with ethyl 3-oxobutanoate and ammonium acetic acid derivation in EtOH gave 1,4-dihydropyridines **54** in 85-93% yield (Scheme 56). The estimation of this technique was quick, straightforward, productive and the recyclability for catalyst. [60].



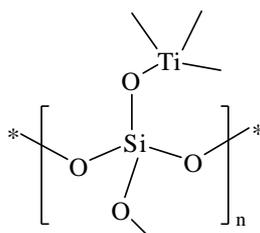
Scheme 56: synthesis of 1,4-dihydropyridines

Preparation of 1,2,4,5-tetra-substituted imidazoles **55** by response of sweet-smelling aldehyde, benzil and amine in the vicinity of ammonium acetic acid derivation utilizing half nano silica upheld titanium chloride (nano-TiCl₄.SiO₂) as a persuasive Lewis corrosive impetus (Scheme 57). This acidic impetus was readied by

means of response of nano SiO₂ with TiCl₄ and was effective, reusable, eco-accommodating and reasonable. [61].

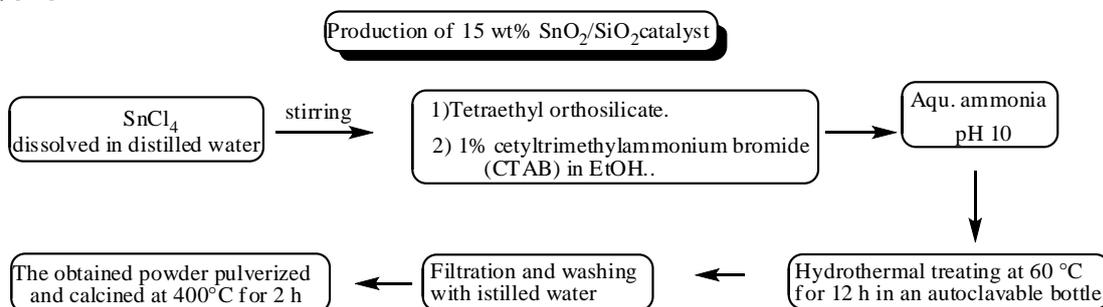


Scheme 57. Synthesis of 1,2,4,5-tetrasubstituted imidazoles

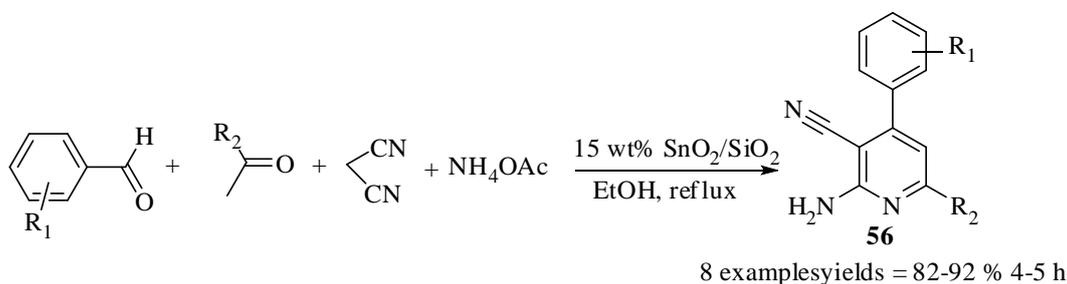


Suggested structure for nano-TiCl₄-SiO₂ Catalyst

2-amino-3-cyanopyridine subordinates **56** were orchestrated in high % yield by means of the cyclo-buildup response of sweet-smelling methyl ketones, aldehydes, malononitrile and ammonium acetic acid derivation catalyzed by SnO₂/SiO₂ nanocomposite [catalysts have been combined by the sol-gel system. creation of 15wt% SnO₂/SiO₂ is arrangements in (Scheme 58)] material at refluxed condition in EtOH (Scheme 59) [62].

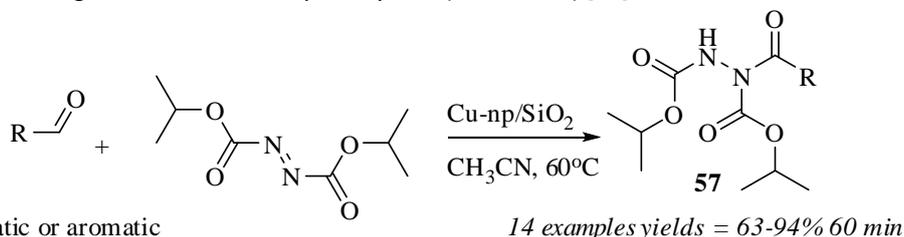


Scheme 58. Preparation of 15wt% SnO₂/SiO₂ catalyst



Scheme 59. Synthesis of 2-amino-3-cyanopyridines

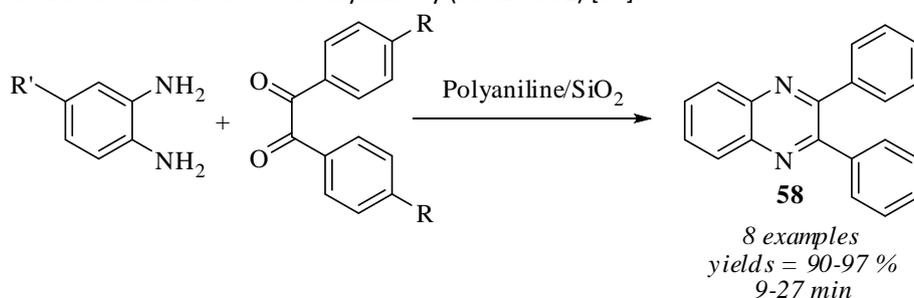
A green, simple and efficient reaction has been accounted for an extensive variety of fragrant and aliphatic aldehydes responded with diisopropyl azodicarboxylate to manage the cost of item 57, (CuO-np/SiO₂) impetus in significant returns. Aliphatic aldehydes gave preferable yields over fragrant aldehydes, this translated by the impact of the electronic properties of the substrates. Likewise, it was seen that electron pulling back gatherings diminished the response yields (Scheme 60) [63].



R=aliphatic or aromatic

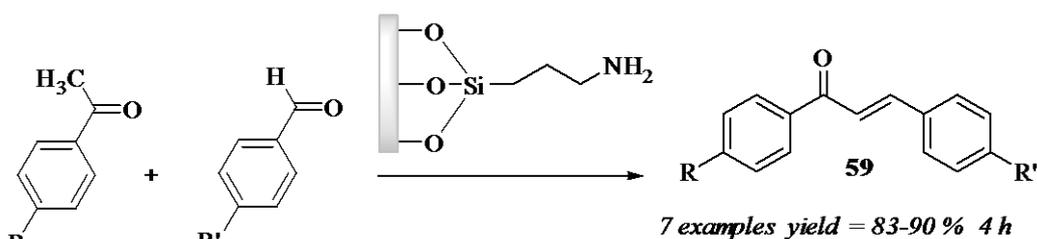
Scheme 60. Synthesis of diisopropyl 1-substituted-benzoylhydrazine-1,2-dicarboxylate

Synthesis of quinoxalines **58** using Polyaniline/SiO₂ nanocomposite as an efficient catalyst at room temperature. The optimal reaction condition done by using 10% catalyst and the catalyst performance was found to be consistent even after three recyclability (Scheme 61) [64].



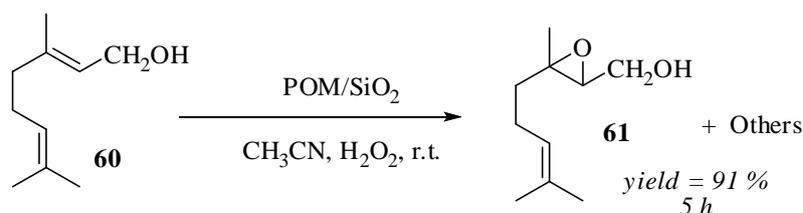
Scheme 61: Synthesis of quinoxaline analogue

Aminopropylated nanosilica was prepared by a simple sol-gel process from tetraethyl orthosilicate (TEOS) and then it was functionalized with different amounts of 3-aminopropyltriethoxysilane (APS) under toluene reflux. The materials were used as catalyst in the Claisen-Schmidt preparation of chalcones **59** for the reaction of substituted acetophenones and benzaldehydes under solvent-free conditions (Scheme 62) [65].



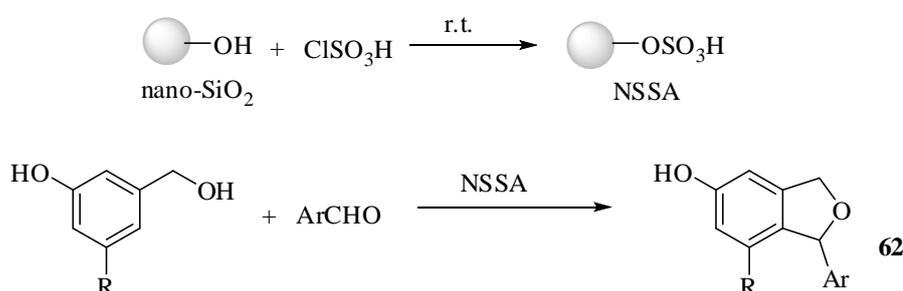
Scheme 62: Claisen-Schmidt chalcones preparation using aminopropylated nanosilica catalyst

Silica nanoparticles supporting polyoxometalates (POMs), in particular an iron (III) mono-substituted Keggin-sort polyoxotungstate of equation $\alpha\text{-[PW}_{11}\text{Fe}^{\text{III}}(\text{H}_2\text{O})\text{O}_{39}]^{4-}$ and a sandwich-sort tungstophosphate with the recipe $\text{B-}\alpha\text{-[PW}_9\text{O}_{34}\text{]}_2\text{Fe}^{\text{III}}_4(\text{H}_2\text{O})_2]^{6-}$ were combined. The POM/SiO₂ nanocomposites were acquired by soluble hydrolysis of tetraethoxysilane utilizing a converse micelle and sol-gel strategy. The synergist action of POM/SiO₂ nanomaterials was tried in the epoxidation of geraniol **60** utilizing H₂O₂ as oxygen giver. The $\alpha\text{-[PW}_{11}\text{Fe}^{\text{III}}(\text{H}_2\text{O})\text{O}_{39}]^{4-}$ /SiO₂ nanocomposite was the most productive impetus with high geraniol change and great regioselectivity for 2,3-epoxygeraniol **62** (Scheme 63)[66].



Scheme 63: Regioselectivity epoxidation of geraniol using POMs catalyst

A simple and efficient synthesis of 3-hydroxyphthalans via oxa-Pictet–Spengler reaction catalyzed by nanosilica sulfuric acid Nanosilica sulfuric acid NSSA was found to be a new, powerful and reusable heterogeneous catalyst for the synthesis of 3-hydroxyphthalans via condensation of 3-hydroxybenzyl alcohols and aromatic aldehydes under heating and microwave irradiation. The reaction has been done by two reaction conditions one of them is stirring vigorously at 80 °C and the other does under microwave irradiation at 80 °C (400 W) (Scheme 64) [67].



*18 examples: Method a yield=55-94 % min 20-90 ,
Method b yield=82-97 % min 90-300*

Scheme 64: Synthesis of 3-hydroxyphthalans catalyzed by NSSA under conventional heating and microwave irradiation

ABBREVIATIONS

SNPs:	Silica nanoparticles
MSNs:	Mesoporous silica nanoparticles
HMF:	Hydroxymethylfurfural
IL:	Ionic liquids
PMOs:	Periodic mesoporous organosilicas
MRNCs:	Magnetically recyclable nano-catalysts
MCM-41:	Mobil Crystalline Material-41
SSA:	Stem Sweep Ash
SBA:	Silica bonded acid
SBNPSA:	Silica-bonded N-propyl sulfamic acid
RHA:	Rice Husk Ash
CPTES:	3-ChloroPropylTriEthoxy-Silane
PW:	12-Tungstophosphoric Acid H ₃ PW ₁₂ O ₄₀
Blm:	1-Butylimidazole
PdNPs:	Palladium Nanoparticles
PEI:	Polyethyleneimine
SPIONs:	SuperParamagnetic Iron Oxide Nanoparticles
MNPs-PSA:	Magnetic Nanoparticles-supported N-propyl-sulfamic
TEOS:	Tetraethyl orthoSilicate
TPI:	Triethoxysilyl-Propyl-Isonicotinamide
HPA/TPI-Fe ₃ O ₄ :	H ₅ PW ₁₀ V ₂ O ₄₀ /pyridino-Fe ₃ O ₄
SEM:	Scanning Electron Microscope
PSNP-CA:	Porous Silica Nano Particle Catalyst
2-TBHQ:	2-Tert-ButylhydroQuinone
APS	3-Aminopropyltriethoxysilane

POMs Polyoxometalates
NSSA Nano silica sulfuric acid

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