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A magnetic palladium nickel carbon nanocomposite as a heterogeneous catalyst for the synthesis of distyrylbenzene and biphenyl derivatives[†]

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A magnetic palladium nickel carbon (Fe₃O₄@Pd@Ni/C) nanocomposite has been synthesized using a simple one-pot procedure *via* a hydrothermal approach. Ferric nitrate, palladium acetate, and nickel nitrate were dissolved in water together with glucose, and the mixture was heated in an autoclave. The Fe₃O₄@Pd@Ni/C nanocomposite was characterized *via* XRD, TEM, FE-SEM, VSM, EDS, and XPS studies. The catalytic abilities of the Fe₃O₄@Pd@Ni/C nanocomposite were investigated for the synthesis of distyrylbenzene and 9,10-distyrylanthracene derivatives. This method shows obvious advantages, such as the recyclability of the catalyst, simple experimental operation, and the obtaining of good to excellent yields.

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Introduction

In recent years, the number of studies focusing on the development and synthesis of multi-metallic nanoparticles has increased.1 Generally, they have been prepared for use as catalysts due to their enhanced properties, such as physical and chemical stability, high selectivity for the target materials, and catalytic activity, when compared with equivalent catalysts based on single metals.¹⁻⁸ Moreover, during preparation, they have been designed with capping agents to allow control of the shape, size, and crystal structure.^{9–12} Among the various hybrid multi-metallic nanoparticles, those hybrids containing iron oxide (Fe₃O₄) have attracted much attention owing to their magnetic recoverability.^{1,6-9} Most prepared hybrid multimetallic nanoparticles have been applied as catalysts in C-C bond formation.¹⁻⁸ The application of palladium species as catalysts in C-C coupling reactions is well known, and it has received considerable attention in both industrial and academic research.¹³⁻¹⁸ Therefore, multi-metallic nanoparticles containing palladium and iron oxide have attracted more attention due to the high catalytic performance of palladium and the magnetic recoverability of Fe₃O₄.¹⁹⁻²⁵

Nickel is a metal that has been recently used as a catalyst in C–C bond formation and organic transformations. In most of these coupling or transformation reactions, 2,4,6-trichloro-1,3,5-triazine (TCT) has been used as an efficient and mild reagent accompanied by nickel-catalyzed species.^{25–30}

These reactions include the amination of phenols,²⁷ the deoxygenation and reductive homocoupling of phenols,²⁸ the conversion of protected phenols into olefins,²⁹ and the reductive amidation of aryl-triazine ethers.³⁰

Conjugated organic materials, those constructed from π -conjugated molecules, have received increasing attention owing to their potential applications in the field of optoelectronics. Recently, 1,4-distyrylbenzene and 9,10-distyrylanthracene derivatives have received increased attention due to their organic-based photoluminescence, organic light-emitting diode (OLED), fluorescence, and electroluminescence applications and their photophysical properties.^{31–38}

Herein we report the preparation of the magnetic nanocomposite Fe_3O_4 @Pd@Ni/C (Scheme 1) as a new heterogeneous catalyst for the synthesis of 1,4-distyrylbenzene, 9,10-distyrylanthracene, and biphenyl derivatives.

Experimental

General

All reagents were purchased from the Merck and Aldrich Chemical Companies. Reaction monitoring was accomplished using TLC on silica gel (PolyGram SILG/UV254) plates. Melting points



Scheme 1 The preparation of the $Fe_3O_4@Pd@Ni/C$ nanocomposite.

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were determined in open capillary tubes using Barnstead Electrothermal 9100 BZ circulating oil melting point apparatus. The compounds were visualized using ¹H-NMR (400 MHz) and ¹³C NMR (100 MHz) spectroscopy, and spectra were recorded using a Bruker Avance instrument in CDCl₃ or DMSO- d_6 solvent with tetramethylsilane (TMS) as an internal standard. FT-IR spectroscopy (Shimadzu FT-IR 8300 spectrophotometer) was employed for the characterization of the products. X-ray diffraction (XRD) data were obtained using a Bruker D8 Advance Theta–2theta diffractometer. Energy dispersive X-ray (EDX) spectroscopy (SiriusSD, England) was used for catalyst analysis. All of the products are known compounds and were characterized *via* ¹H and ¹³C NMR spectra (see the ESI†) and compared with the literature.^{1,32,33,36,39}

Preparation of palladium nickel ferrite (Fe₃O₄@Pd@Ni/C nanocomposite)

Fe(NO₃)₃·9H₂O solution (1 M, 2 equiv.), Pd(OAc)₂ solution (1 M, 0.5 equiv.), and Ni(NO₃)₂·6H₂O solution (1 M, 0.5 equiv.) were dispersed in 10 mL of distilled water and added to a mixture containing glucose solution (1 M). After being stirred for about 30 min, the mixture was transferred into a Teflon-lined stainless-steel autoclave. The autoclave was maintained at 180 °C for 24 h and then naturally cooled to ambient temperature. The precipitate was filtered, washed with water and ethanol, and dried at 100 °C. Finally, the nanocomposite was obtained after keeping the temperature at 250 °C for 3 h. The synthesized Fe₃O₄@Pd@Ni/C nanocomposite was characterized using different methods, including SEM, TEM, IR, XRD, EDS, and XPS.

General procedure for the synthesis of distyrylbenzenes

The Fe₃O₄@Pd@Ni/C catalyst (0.075 g, 3.29 mol% with respect to the Pd content) was added to a mixture of aryl halide (1 mmol), alkene (1.2 mmol), and Na₂CO₃ (2.5 mmol) in DMF (3 mL), and this was heated in an oil bath at 120 °C. The reaction was continuously followed *via* TLC to completion. After completion, the catalyst was removed using an external magnet and was washed with dichloromethane (3 × 5 mL). Having extracted the reaction mixture from dichloromethane and water, the organic phase was dried over Na₂SO₄. The evaporation of the solvent gave the products. For further purification, the products were passed through a short column of silica gel using *n*-hexane as the eluent.

[It is worth mentioning that in the cases of 1,4dihalobenzene and 9,10-dibromonanthracene (1 mmol), the following conditions were used: alkene (2.4 mmol) and Na₂CO₃ (2.5 mmol) in DMF (3 mL); Fe₃O₄@Pd@Ni/C catalyst (0.15 g, 6.58 mol% with respect to the Pd content); oil bath at 120 °C].

General procedure for the synthesis of biphenyls

The Fe₃O₄@Pd@Ni/C catalyst (0.05 g, 2.19 mol% with respect to the Pd content) was added to a round-bottom flask (10 mL) containing aryl halide (1 mmol), phenylboronic acid derivative (1.2 mmol), and sodium carbonate (2.5 mmol) in water (3 mL), and this was heated in an oil bath at 80 °C. The reaction was continuously followed using TLC to completion. After completion, the catalyst was removed using an external magnet and was washed with dichloromethane (3 \times 5 mL). Having extracted the reaction mixture from dichloromethane and water, the organic phase was dried over Na₂SO₄. The evaporation of the solvent gave the products. For further purification, the products were passed through a short column of silica gel using *n*-hexane as the eluent.

[It is worth mentioning that in the case of 1,4-dihalobenzene (1 mmol), the following conditions were used: phenylboronic acid derivative (2.4 mmol) and Na_2CO_3 (2.5 mmol) in water (3 mL); Fe₃O₄@Pd@Ni/C catalyst (0.1 g, 4.38 mol% with respect to the Pd content); oil bath at 80 °C].

Results and discussion

Initially, the magnetic palladium nickel carbon (Fe_3O_4 @Pd@ Ni/C) nanocomposite was prepared *via* the reaction of iron(III) nitrate, palladium acetate, and nickel nitrate in deionized water in the presence of glucose. The structure and physical and chemical properties of the double salt were investigated using FT-IR, EDS, XRD, XPS, SEM, and TEM methods.

FT-IR spectra of the prepared nanocomposite are presented in Fig. 1. FT-IR analysis of the nanocomposite (Fe₃O₄@Pd@Ni/C) shows characteristic peaks at around 449 cm⁻¹ and 576 cm⁻¹, which can be assigned to the presence of metal-oxide (Fe–O and Ni–O) vibrations. The FT-IR spectrum of the recovered nanocomposite showed no obvious changes in comparison with the freshly synthesized catalyst (Fig. 1b).

The morphology of the nanocomposite (Fe_3O_4 @Pd@Ni/C) was investigated *via* SEM and TEM methods. SEM images demonstrate that the obtained nanocomposite has a uniform structure, and aggregate spheres are detected on the carbon backbone (Fig. 2). TEM and high-resolution TEM images are shown in Fig. 3. The TEM images clearly revealed the uniform sphere-shaped morphology with an obvious lattice, and the corresponding HR-TEM images also confirmed the crystallite size of the synthesized nano-composite (PdNiFe₃O₄). The diameter of the nano-magnetic (Fe_3O_4) core was around 388 nm and TEM images of the catalyst show Pd and Ni nanoparticles with near-spherical morphology, which were aggregated on the nano-magnetic core. Also, the obtained histogram (Fig. 4) confirmed



Fig. 1 $\,$ FT-IR spectra of the nanocomposite: (a) fresh Fe_3O_4@Pd@Ni/C and (b) the catalyst after recovery.



Fig. 2 SEM images of the nanocomposite ($Fe_3O_4@Pd@Ni/C$).

that the size distribution of the nanoparticles in this nanocomposite was narrow and normal, with average values between 80 and 90 nm.

The EDX pattern of the nanocomposite (Fe₃O₄@Pd@Ni/C) is shown in Fig. 5. This analysis indicated the presence of the constituent elements Pd, Fe, Ni, O, N, and C in the nanocomposite. The C, Fe, Ni, and Pd content levels in Fe₃O₄@Pd@Ni/C were determined to be 8.02%, 30.69%, 30.66%, and 12.38% (w/w), respectively.

The structure of the synthesized magnetic palladium nickel carbon nanocomposite (Fe₃O₄@Pd@Ni/C) was studied *via* XRD analysis.

Fig. 6 shows the XRD pattern of this nanocomposite. As can be seen from the XRD spectrum of the nanocomposite, the diffraction peaks at 2θ values of 18.7, 30.4, 35.7, 43.4, 54.1, 57.6, 63.1, and 68.3° can be attributed to Fe₃O₄. Moreover, the diffraction peaks at 2θ values of 34.1, 37.5, and 60.3° correspond to nickel. The diffraction peaks at 2θ values of 40.4, 46.8, and 68.3° correspond to the Pd planes of the nanocomposite.¹



Fig. 3 TEM (a) and HR-TEM (b) images of the nanocomposite (Fe3O4@Pd@Ni/C).



Fig. 4 A histogram showing the nanoparticle size distribution in the nanocomposite.



Fig. 5 EDX spectra of (a) the fresh nanocomposite (Fe $_3O_4$ @Pd@Ni/C) and (b) the recovered material after eight cycles.

Through a comparison with the XRD patterns of the constituent components (palladium, nickel oxide, and nickel ferrite), it is indicated that the nanocomposite is made up of these elements.

Magnetic hysteresis measurements of Fe_3O_4 (a)Pd (a)Ni/C were done using an applied magnetic field at r.t., with the field being



Fig. 6 Top panel: the XRD patterns of (a) fresh $Fe_3O_4@Pd@Ni/C$ and (b) the material after recovery. Bottom panel: the XRD peaks of the nanocomposite ($Fe_3O_4@Pd@Ni/C$) compared with the standard patterns of Pd, NiO, and NiFe_2O_4.

swept from $-15\,000$ to $15\,000$ Oersted. As shown in Fig. 7, these nanoparticles exhibited superparamagnetic characteristics, as the M (H) hysteresis loop for the sample was completely reversible. The hysteresis loop reached saturation at the maximum applied magnetic field value. The magnetic saturation value of the catalyst was 18 emu g⁻¹ at r.t.

X-ray photoelectron spectroscopy (XPS) analysis was carried out to further measure the chemical composition of Fe_3O_4 @Pd@Ni/C. The survey spectrum of Fe_3O_4 @Pd@Ni/C displays that the nanocomposite consists of C, N, O, Pd, Ni, and Fe elements, with atomic percentages of 72.78%, 6.74%,



Fig. 7 The VSM spectrum of the nanocomposite (Fe₃O₄@Pd@Ni/C).

16.54%, 0.86%, 1.31%, and 1.77%, respectively (Fig. 8a). As displayed in Fig. 8b, the peaks at 284.32 eV, 284.93 eV, 286.1 eV, 287.16 eV, and 288.8 eV can be attributed to carbon in C-C/C=C, C-N, C-O, C=O, and O-C=O bonds, respectively.

In Fig. 8c, the peaks located at 399.10 eV, 399.95 eV, 400.89 eV, and 401.76 eV can be assigned to nitrogen in N-pyridinic, N-pyrrolic, N-graphitic, and N-oxidised forms, respectively. The Pd XPS spectrum in Fig. 8d shows a double peak at binding energies (BEs) of 335.34 and 340.78 eV, corresponding to Pd $3d_{5/2}$ and Pd $3d_{3/2}$, respectively. The BEs of the doublet for Pd $3d_{5/2}$ and Pd $3d_{3/2}$ are characteristic of Pd(0). In addition, the double Pd $3d_{5/2}$ and Pd $3d_{3/2}$ peaks at 336.15 and 341.98 eV, and 337.16 and 342.88 eV correspond to PdO and Pd(OH)_x, respectively. The amounts of Pd(0), PdO, and Pd(OH)_x are 2.23%, 2.18%, and 0.92%, respectively.

The Ni XPS spectrum shows $2p_{3/2}$ and $2p_{1/2}$ peaks at 855.6 and 872.98 eV, and 856.78 and 873.88 eV, as shown in Fig. 8e, which are characteristic of Ni²⁺ and Ni³⁺, respectively. This indicates that the Ni nanoparticles were oxidized owing to exposure to air or under reaction conditions during the preparation of the catalyst. Furthermore, Fig. 8f shows the XPS signals from the Fe 2p region, which indicate that Fe²⁺ and Fe³⁺ formed Fe₃O₄. Fe $2p_{3/2}$ and Fe $2p_{1/2}$ peaks are seen at 710.23 and 723.28 eV, and 711.88 and 725.08 eV, and these could be attributed to Fe²⁺ and Fe³⁺, respectively.

The ICP data show 4.67% w/w palladium in Fe₃O₄@Pd@Ni/ C. Because of the Pd content, the nanocomposite catalyst was treated with concentrated HCl and HNO₃ to digest the Pd species, followed by ICP analysis. The Pd, Ni, and Fe content levels in Fe₃O₄@Pd@Ni/C were determined to be 4.67%, 41.38%, and 35.14% w/w, respectively. Here, 0.075 g of catalyst is used per 1 mmol of aryl-halide, which is equivalent to 0.0035 g of Pd metal, corresponding to 3.29 mol% catalyst per 1 mmol of aryl-halide used.

After the preparation and characterization of the Fe_3O_4 (a)Pd(a) Ni/C nanocomposite, its catalytic activity was investigated for C–C bond formation *via* the Heck reaction and Suzuki coupling reaction.

Initially, the carbon–carbon coupling reaction between iodobenzene and ethyl acrylate was selected as a model reaction, and the reaction time and yield were measured in the presence of the Fe_3O_4 @Pd@Ni/C nanocomposite while varying reaction parameters such as the solvent, catalyst loading, base, and reaction temperature (Scheme 2 and Table 1).

As shown in Table 1, the model reaction was not accomplished in the absence of catalyst, even after 24 h at 120 $^{\circ}$ C (Table 1, entry 1).

The model reaction was carried out in the presence of palladium acetate (0.01 g, 9 mol% Pd), and the corresponding product was obtained in 84% yield after 12 h at 120 $^{\circ}$ C (Table 1, entry 2).

The model reaction was carried out in the presence of nickel nitrate and did not give any product (Table 1, entry 3). The best results were achieved in the presence of the Fe₃O₄@Pd@Ni/C nanocomposite (0.075 g, 3.29 mol% Pd) in DMF (3 mL) with sodium carbonate (2.5 mmol) at 120 °C. Decreasing the amount



Fig. 8 (a) The XPS survey spectrum of the Fe₃O₄@Pd@Ni/C nanocomposite, and high-resolution (b) C 1s, (c) N 1s, (d) Pd 3d, (e) Ni 2p, and (f) Fe 2p XPS spectra.



of Fe₃O₄@Pd@Ni/C nanocomposite led to a decrease in the product yield (Table 1, entries 4 and 5). The model reaction was also carried out in the presence of other bases, including K_2CO_3 , Na_3PO_4 , and Et_3N in DMF at 120 °C, and Na_2CO_3 was the best base for this reaction (Table 1, entries 4–10). In another effort, the model reaction carried out at lower temperatures with DMF as the solvent gave the corresponding product in lower yield (Table 1, entry 16).

The reaction was studied in the presence of the Fe_3O_4 Pd@Ni/C nanocomposite in various solvents, such as toluene, dioxane, water, acetonitrile, and dimethylacetamide (DMA), and the reaction proceeded in DMF and DMA solvents (Table 1, entries 11–15).

Since 1,4-distyrylbenzenes and 9,10-distyrylanthracenes have photoelectronic and fluorescence properties,¹⁻⁶ [1-6], in this study we have focused on the synthesis of series of these compounds using the magnetic palladium nickel nanocomposite as a new catalyst. For this purpose, we used the Heck reactions shown in Schemes 3 and 4.

As shown in Schemes 5, 1,4-diiodobenzene (4) reacted with ethyl acrylate or methyl acrylate (2) or styrene derivatives (5) under the optimized conditions, and the corresponding products (**6a–6c** and **7a–7c**) were obtained with yields in the range of 72–86%. Moreover, when 2,5-dimethyl-1,4-diiodobenzene and

Table 1 Optimization of the reaction conditions^a

Entry	Solvent	Base	Catalyst loading (g)	Temp. (°C)	Time (h)	Yield ^b (%)
1	DMF	Na ₂ CO ₃	_	120	24	0
2	DMF	Na ₂ CO ₃	$0.01 (9 \text{ mol}\%)^c$	120	12	84
3	DMF	Na ₂ CO ₃	$0.01 (1.1 \text{ mol}\%)^d$	120	12	0
4	DMF	Na ₂ CO ₃	0.025	120	8	70
5	DMF	Na ₂ CO ₃	0.05	120	8	83
6	DMF	Na ₂ CO ₃	0.075	120	8	90
7	DMF	Na ₂ CO ₃	0.1	120	8	90
8	DMF	K_2CO_3	0.075	120	8	85
9	DMF	Na_3PO_4	0.075	120	8	85
10	DMF	Et ₃ N	0.075	120	8	54
11	Toluene	Na ₂ CO ₃	0.075	Reflux	8	25
12	Dioxane	Na_2CO_3	0.075	Reflux	8	Trace
13	Acetonitrile	Na_2CO_3	0.075	Reflux	8	41
14	Water	Na_2CO_3	0.075	Reflux	8	10
15	DMA	Na ₂ CO ₃	0.075	120	8	90
16	DMF	Na ₂ CO ₃	0.075	80	8	56
17	DMF	Na ₂ CO ₃	0.075	140	8	91

^a Reaction conditions: iodobenzene (1 mmol), ethyl acrylate (1.2 mmol), and base (2.5 mmol) in solvent (3 mL). ^{*b*} Isolated yield. ^{*c*} $Pd(OAc)_2$ was used as the catalyst. ^{*d*} $Ni(NO_3)_2$ was used as the catalyst.

2,5-dimethoxy-1,4-diiodobenzene as ortho-substituted aryl halides reacted with ethyl acrylate or styrene, the corresponding product 6c or 7c was obtained in 83% yield after 8 h or 71% yield after 12 h, respectively. In addition, when 1,4-dibromobenzene was used as the aryl halide, it gave the corresponding products after longer reaction times (24 h) and in lower yields (46-62% yields). Also, 1,4dichlorobenzene was treated with ethyl acrylate (2) under optimized conditions, but after 36 h this gave only a trace of the product.

Subsequently, 9,10-dibromoanthracene (8) was reacted with ethyl acrylate (2) and styrene derivatives (5) under the optimized conditions, giving the corresponding products 9 and 10a-10c, respectively, in good to excellent yields (Scheme 6).

Encouraged by the obtained results from the Heck reaction, we also investigated the potential of our Fe₃O₄@Pd@Ni/C catalyst for use in the Suzuki cross-coupling reaction. First, a coupling reaction between 4-iodoanisole and phenylboronic acid was chosen as a model reaction (Scheme 7). Due to environmental concerns, the reaction was only carried out with water as the solvent and sodium carbonate as the base. Next, we examined the effect of catalyst loading on the yield of the coupling product. Based on the results, the optimized conditions were chosen as follows: water (3 mL) as the solvent; Na₂CO₃ (2.5 mmol) as the base; a catalyst loading of 0.05 g (2.19 mol%



Scheme 3 The synthesis of distyrylbenzenes in the presence of Fe₃O₄@Pd@Ni/C as a catalyst.





Scheme 4 The synthesis of 9,10-distyrylanthracenes in the presence of Fe₃O₄@Pd@Ni/C as a catalyst.



Fe₃O₄@Pd@Ni/C as a catalyst.

based on the Pd content); and heating at 80 °C. 4-Methoxybiphenyl was obtained as the corresponding product (13) after 80 min and in 91% yield. Using lower catalyst amounts gave longer reaction times and lower yields.

Again, in this study, we focused on the reaction of 1,4diiodobenzene with phenylboronic acid derivatives under the optimized conditions, and the obtained results are summarized in Scheme 8. 1,4-Diiodobenzene reacted with aryl boronic acids, including phenyl, 3-methyl-phenyl, 2-methyl-phenyl,

Scheme 6 The synthesis of 9,10-distyrylanthracenes in the presence of Fe₃O₄@Pd@Ni/C as a catalyst





Fig. 9 Images of (a) fresh catalyst powder, (b) the reaction mixture, and (c) the recovery of the catalyst using an external magnet.



Fig. 10 Yields after the recovery of the $Fe_3O_4@Pd@Ni/C$ nanocomposite in the reaction between iodobenzene and ethyl acetoacetate under the optimized conditions (product 3).





Table 2 A comparison of ${\rm Fe_3O_4@Pd@Ni/C}$ with other reported reagents and catalysts

Compound	Catalyst and conditions	Time (h)	Yield (%)	Ref.
7b	Pd(OAc) ₂ , 130 °C in NMP	15	71	35
7 b	Fe ₃ O ₄ @Pd@Ni/C, 120 °C in DMF	12	75	This work
10a	Wittig method	5	75	36
10a	Fe ₃ O ₄ @Pd@Ni/C, 120 °C in DMF	8	87	This work
10b	Pd(OAc) ₂ , 140 °C in DMA	24	73	32
10b	Fe ₃ O ₄ @Pd@Ni/C, 120 °C in DMF	8	85	This work
10c	Wittig method	5	65	36
10c	Fe_3O_4 @Pd@Ni/C, 120 °C in DMF	8	86	This work

4-fluorophenyl, and 1-naphthyl examples, under the optimized conditions in the presence of the Fe_3O_4 (a)Pd (a)Ni/C nanocomposite, giving the corresponding products (**14a–14e**) in good to excellent yields (Scheme 8). It worth mentioning that when 4-methoxyphenyl boronic acid was treated with 1,4-diiodobenzene under the optimized conditions for 90 min, 4-iodo-4'-methoxy-1,1'-biphenyl (**15**) was obtained as the major product (87% yield).

Finally, the recycling of the catalyst was examined after the synthesis of compounds 3 and 13. After the completion of the reactions, the catalyst was removed using an external magnet (Fig. 9).

The obtained catalyst was washed with dichloromethane. The recovered catalyst was successfully reused several times without a significant loss of catalytic activity (Fig. 10 and 11).

To investigate the stability of the recovered catalyst, its structure was studied using methods such as IR, EDX, and XRD. The IR, EDX, and XRD spectra of the recovered nanocomposite showed no obvious changes in comparison with the freshly synthesized catalyst (Fig. 1b, 5b and 6b).

Finally, as shown in Table 2, the advantages and applicability of the Fe_3O_4 @Pd@Ni/C catalyst in the Heck reaction were compared with other reported catalysts and reagents.

Conclusions

In summary, the hydrothermal preparation of a nanocomposite (Fe₃O₄@Pd@Ni/C) was reported. The catalyst was characterized, and it showed efficient abilities to catalyze the Heck and Suzuki

reactions. 1,4-Distyrylbenzene and 9,10-distyrylanthracene derivatives were prepared in the presence of the Fe₃O₄@Pd@Ni/C nanocomposite as a recyclable catalyst in good to excellent yields. The presented methodology is simple to operate, has reasonable reaction times, involves a recoverable catalyst, generates high yields, has an easy work-up procedure (using an external magnet), and uses aqueous media and environmentally benign mild reaction conditions for Suzuki reactions.

Conflicts of interest

There is no conflicts to declare.

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