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Catalytic performance of Cu(II)-supported graphene quantum dots modified NiFe₂O₄ as a proficient nano-catalyst in the synthesis of 1,2,3-triazoles

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Abstract

 $NiFe_2O_4$ nanoparticles are modified by graphene quantum dots (GQDs) and utilized to stabilize the Cu(II) nanoparticles as a novel magnetically retrievable catalytic system (Cu(II)/GQDs/NiFe₂O₄) for green formation of 1,4-disubstituted 1,2,3-triazoles by means of alkyne–aryl azide cycloaddition. The prepared catalyst can be isolated assisted by an outer magnet and recovered for five courses without significant reduction in its efficiency. The as-prepared magnetic heterogeneous nanocomposite was characterized by UV–Vis, FT-IR, XRD, EDS, VSM, TEM, and ICP. Performing the reactions in environmentally friendly and affordable conditions (water), the low catalyst percentage, high yield of products, short reaction times, and easy workup are the merits of this protocol.

Graphic abstract



Keywords NiFe₂O₄ · Graphene quantum dots · 1,2,3-Triazole · Nano-catalyst

Introduction

Graphene quantum dots (GQDs) are the zero-dimension nano-graphenes with special characteristics like small size, chemical inertness, photoluminescence, ease to be functionalized with biomolecules, and biocompatibility have received a lot of attention in the nanotechnology researches [1–3]. Nevertheless, the diverse uses of GQD, little consideration has been given for applying GQD as solid support or catalyst in the reactions [4–6]. Preparation of high-performing nano-catalysts for organic reactions is

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Farid Moeinpour fmoeinpour@iauba.ac.ir remain a major challenging task. To achieve greater specific area and more effective sites, nano-catalysts should be functionalized by activated moieties [7–9]. It is proved that the modification of the nano-catalyst with GQDs avoids the aggregation of fine particles and, therefore, enhances the active specific area for an effective catalytically reaction [8, 9]. Magnetic nanoparticles have been the focus of attention to researchers for consecutive years [10]. Many works have been published and also indicated the importance of magnetic catalysts [11–14]. Among them, $NiFe_2O_4$ has been getting more consideration, owing to its strong coercive power, appropriate magnetic induction, and high permanence [15]. Several researches verify the outlooks of using NiFe₂O₄ as an effective nano-catalyst [16–19]. The immobilization of nano-NiFe₂O₄ on GQDs will lead to a multi-purpose nano-scaffold for efficient catalytic activity [20]. The cycloaddition reaction between alkynes and azides presented by Huisgen [21].

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This reaction without catalyst usually generates two isomeric triazoles (1,4- and 1,5-), making it unsuitable for organic preparations. In 2002, the application of copper to the reaction under mild conditions was described [22–24]. Lately, this procedure has been developed as a prevalent way for preparation of triazoles by diverse catalysts such as copper(II) acetate [25], nano-CuO [26], silica/nano-Cu [27], nanomagnetic/Cu [28], Cu nanoparticles immobilized on modified magnetic zeolite [14], and copper(II) porphyrin graphene oxide [29]. Nevertheless, a large

number of these procedures have impediments that must be considered. Therefore, the application of newer and greener approaches is remaining necessary. Herein, the preparation of Cu(II)/GQDs/NiFe₂O₄ by way of an easy co-sedimentation of NiFe₂O₄ nanoparticles on GQDs and its identification by diverse methods are discussed. The catalyst synthesis process is illustrated in Scheme 1. Upon full identification, the catalyst performance was investigated in preparation of 1,4-disubstituted 1,2,3-triazole derivatives. The overall reaction is shown in Scheme 2.



1a: R = Ph, **1b**: 4-NO₂Ph, **1c**: 4-BrPh, **1d**: 4-CH₃Ph, **1e**: 4-CIPh, **1f**: 4-CH₃OPh, **1g**: PhCH₂ **2a**: R¹ = Ph, **2b**: R¹ = CH₂OH, **2c**: R¹ = CH₂Br

Scheme 1

Result and discussion

The catalyst (Cu(II)/GQDs/NiFe₂O₄) was synthesized as specified by the procedure outlined in Scheme 1, by means of GQDs and NiFe₂O₄ nanoparticles produced in accord with literature [6, 15, 20, 30]. To verify synthesis of the nano-catalyst, it was completely identified using diverse methods comprising UV–Vis and fluorescence spectroscopy, FT-IR spectroscopy, XRD, EDS, TEM, VSM, and ICP spectroscopy.

The light-conducting characteristics of the prepared graphene quantum dots were examined by UV–Vis and fluorescence spectroscopy. The absorbing spectrum of graphene quantum dots does not give any peak as illustrated in Fig. 1a. The fluorescence spectra of graphene quantum dots could be observed in Fig. 1b. The graphene quantum dots were excited at wavelengths of 360, 380, and 400 nm and



Fig.1 a UV–Vis absorbance spectrum of GQDs. **b** Fluorescence spectra of GQDs. Inset indicates images of GQDs solution taken under visible light (weak yellow) and 365 nm excitation (blue) (color figure online)

the proper fluorescence emission peaks were found at 454, 463, and 469 nm respectively. The emission peaks indicate a gentle red shift with a raising in the excitation wavelengths (360-400 nm). It has been observed that the color of the GQDs aqueous solution is weak yellow under visible light, but when excited by UV light at 365 nm, it seems blue, as indicated in the Fig. 1b. Changes in fluorescence intensity after the synthesis of GQDs/NiFe2O4 were also examined. The fluorescence spectra of GQDs/NiFe₂O₄, excited at 360, 380, and 400 nm is indicated in Fig. 2. The fluorescence emission peaks were found at 486, 483, and 480 nm, respectively. It has been found that with increasing excitation wavelengths, a slight blue shift is observed in emission peaks. This differentiation in the fluorescence properties in comparison to the graphene quantum dots demonstrate the change in the chemical surface of the GQDs/NiFe₂O₄ implying a probable linkage of graphene quantum dots on the nickel ferrite nanoparticles, leading to a change in the surface characteristics of graphene quantum dots.

FT-IR spectroscopy was accomplished to validate the exterior framework of the nano-catalyst. The FT-IR spectra of NiFe₂O₄ and Cu(II)/GODs/NiFe₂O₄ have been displayed in Fig. 3. The wide peak at 3362.97 cm^{-1} is assigned to the hydroxyl (O-H) groups on the basal plane of GQDs and also showing the absorption of water by the Cu(II)/GODs/ NiFe₂O₄. The bands located at 2981 cm⁻¹ and 1450 cm⁻¹ are related to the C-H bond vibrational stretching from C-H bonds on the basal plan of GQDs and remaining citric acid, implying the partial citric acid carbonization [31]. The peak located at 1001.35 cm⁻¹ is related to the vibrational stretching of the C-O-C bond. The peaks located at 1415.34 and 1594.81 cm⁻¹ would be the result of skeletal vibrations of aromatic rings in graphene quantum dots [32]. The presence of NiFe₂O₄ is validated by absorption bands located at 550.64 and 685.24 cm⁻¹, which are correlated to Ni–O and Fe–O bonds vibrations, respectively [33]. The outcomes



Fig. 2 Fluorescence spectra of Cu(II)/GQDs/NiFe₂O₄



Fig. 3 FT-IR spectra of a NiFe₂O₄ and b Cu(II)/GQDs/NiFe₂O₄

of this analysis confirm successful synthesis of magnetic nano-catalyst.

X-ray diffraction technique was employed to acquire information about the crystallinity of the nano-catalyst. XRD pattern of GQDs/NiFe₂O₄ was displayed in Fig. 4. The GQDs/NiFe₂O₄ diffraction pattern was agreed well with the standard NiFe₂O₄ (JCPDS 10-0325) exhibits the important diffraction lines at $2\theta = 17.15^{\circ}$, 30.59° , 35.76° , 37.30° , 43.43° , 53.84° , 57.34° , and 63.01° , which may be attributed the (220), (311), (222), (400), (422), (511), and (440) planes of NiFe₂O₄, respectively. The diffraction peak of graphene quantum dots (004) (JCPDS 26-1080) is not recognizable, may have been due to their high dispersals R. Deilam et al.

and low crystallization degree of graphene quantum dots in $GQDs/NiFe_2O_4$ [15, 34].

The EDS was employed as an influential method to identify the chemical constitution of the produced nano-catalyst. The EDS analysis verifies the existence of envisaged elements comprising nickel, iron, oxygen, carbon, and copper in the catalyst structure (Fig. 5).

The morphology and structural characteristics of the synthesized nano-catalyst were observed under TEM technique (Fig. 6). It is revealed that the darker region in Cu(II)/GQDs/NiFe₂O₄ image, is related to agglomeration of NiFe₂O₄ nanoparticles on GQDs. Also, the TEM micrograph of the nano-catalyst indicates that the mean sizes of Cu(II)/GQDs/NiFe₂O₄ nanoparticles are approximately not more than 40 nm. The specified area of the Cu(II)/GQDs/NiFe₂O₄ catalyst been tested by BET and was 421.2 m²/g.

The magnetic characteristic of Cu(II)/GQDs/NiFe₂O₄ was studied by VSM. As evidenced in Fig. 7, the value of the saturation magnetization of Cu(II)/GQDs/NiFe₂O₄ (60.82 emu g⁻¹), almost equal to Fe₃O₄ nanoparticles (61.60 emu g⁻¹) which indicates that the nano-catalyst has magnetic properties and their magnetic characteristics are so high that they could be isolated by a typical magnet.

The quantity of Cu loading onto the nano-catalyst was determined by the ICP technique which was obtained to be 0.94 mmol g^{-1} .

After validating the successful synthesis of nano-catalyst by various techniques, its catalytic capability was examined in the synthesis of 1,2,3-triazoles. To achieve this goal, 4-nitrophenyl azide and phenylacetylene were used as model substrates for optimizing the reaction elements including solvent, nano-catalyst amount, and temperature. At the beginning, the above reaction was also conducted without and in the presence of various quantities of the nano-catalyst (Table 1). The outcomes revealed that the reaction did not progress in the absence of nano-catalyst in some polar and



Fig. 4 XRD pattern of Cu(II)/ GQDs/NiFe₂O₄



Fig. 5 EDS pattern of Cu(II)/GQDs/NiFe₂O₄





Fig. 7 Magnetization curve of Cu(II)/GQDs/NiFe₂O₄

Fig. 6 TEM image of $Cu(II)/GQDs/NiFe_2O_4$

non-polar solvents and solvent-free state, even after 24 h (entries 1–6). Enhancing the nano-catalyst dosage to 10 mg raised the reaction efficiency (entries 7–14). Moreover, the reaction yield in polar solvents was better than those of non-polar solvents. Lastly, the greatest efficiency was achieved when the model reaction was performed in a H₂O in the presence of 10 mg of nano-catalyst at 60 °C (entry 15). To illustrate the role of copper in the reaction progress, the function of other components of the nano-catalyst like NiFe₂O₄, GQDs, and GQDs/NiFe₂O₄ was also examined in the reaction under optimal conditions. The outcomes are displayed in Table 1 (entries 17–19). As illustrated, no

progress was detected in the model reaction when applying the other components of nano-catalyst in the absence of copper, verifying that the existence of copper is vital for catalyzing the reaction. Also, the model reaction was carried out using copper(II) acetate (Cu(OAc)₂) for 1 h at 60 °C, 1-(4-nitrophenyl)-4-phenyl-1*H*-1,2,3-triazole was obtained in 21% isolated yield (entry 20). The result shows that Cu(OAc)₂ displayed poor activity under optimal conditions, in contrast to those with Cu(II)/GQDs/NiFe₂O₄.

After creating optimal reaction conditions, $Cu(II)/GQDs/NiFe_2O_4$ the range of the reaction was expanded to diverse azides and various terminal alkynes. In accord with the outcomes presented in Table 2, the electronic effects did not have extraordinary affection on outcome of the reaction, thus

Entry	Condition Catalyst /mg, m	nol%	Temp. /°C	Time /h	Yield /% ^a	TON ^b	TOF ^c
1	H ₂ O	_	rt	24	Trace	_	_
2	MeOH	-	rt	24	Trace	-	-
3	EtOH	-	rt	24	Trace	-	-
4	<i>n</i> -Hexane	_	rt	24	Trace	_	-
5	Solvent-free	-	rt	24	Trace	-	-
6	H ₂ O/EtOH	-	rt	24	Trace	-	-
7	MeOH (Cu(II)/GQDs/NiFe ₂ O ₄)	5.00, 0.47	rt	5	12	25.53	5.11
8	EtOH (Cu(II)/GQDs/NiFe2O4)	5.00, 0.47	rt	5	15	31.91	6.38
9	<i>n</i> -Hexane (Cu(II)/GQDs/ NiFe ₂ O ₄)	5.00, 0.47	rt	5	Trace	-	-
10	Solvent-free (Cu(II)/GQDs/ NiFe ₂ O ₄)	5.00, 0.47	rt	5	Trace	-	-
11	H ₂ O/EtOH (Cu(II)/GQDs/ NiFe ₂ O ₄)	5.00, 0.47	rt	5	18	38.30	7.66
12	H ₂ O (Cu(II)/GQDs/NiFe ₂ O ₄)	5.00, 0.47	rt	5	30	63.83	12.77
13	H ₂ O (Cu(II)/GQDs/NiFe ₂ O ₄)	10.00, 0.94	40	2.5	40	42.55	17.02
14	H ₂ O (Cu(II)/GQDs/NiFe ₂ O ₄)	10.00, 0.94	50	2.5	60	63.83	25.53
15	H ₂ O (Cu(II)/GQDs/NiFe ₂ O ₄)	10.00, 0.94	60	0.25	98	104.26	417.02
16	H ₂ O (Cu(II)/GQDs/NiFe ₂ O ₄)	10.00, 0.94	80	0.25	98	104.26	417.02
17	H ₂ O (NiFe ₂ O ₄)	10.00, 0.94	60	1.0	None	_	-
18	H ₂ O (GQDs)	10.00, 0.94	60	1.0	None	-	-
19	H ₂ O (GQDs/NiFe ₂ O ₄)	10.00, 0.94	60	1.0	None	_	-
20	Cu(OAc) ₂	10.00, 0.94	60	1.0	21	22.34	22.34

Table 1 Optimization of solvent, temperature, and nano-catalyst dosage

^aOn the basis of isolated yield

^bTurnover number

^cTurnover frequency = TON/time

all aryl azides with electron-withdrawing or electron-releasing groups provided similar yields (7–28 min). Benzyl azide in comparison to phenyl azides needed shorter time for the reaction (entries 13–15). It seems that the greater electronreleasing effects of the benzyl group (Ph-CH₂), compared to the phenyl group, increase the electron density on nitrogen atoms in benzyl azides and enhance their tendency to attack phenyl acetylene. Each of the aromatic and aliphatic terminal acetylenes produced the related 1,2,3-triazoles in excellent efficiencies.

In accord with literature [35, 36], a plausible catalytic mechanism for the preparation of 1,2,3-triazoles by Cu(II)/ GQDs/NiFe₂O₄ is shown in Scheme 3. At first, alkyne is coordinated to Cu of the nano-catalyst. This interaction speeds up activation of the C–H bond and thereby facilitates the copper–alkylidine complex formation. Next step includes reaction of aryl azide with the Cu–alkylidine intermediate, accompanied by intramolecular cyclization to generate the five-membered triazole ring.

The reusability of the catalyst $(Cu(II)/GQDs/NiFe_2O_4)$ was examined in the sample reaction. For this goal, upon termination of the reaction, the nano-catalyst was separated with the aid of a magnet and washed with acetone and water

to eliminate remained product, dried and reapplied in subsequent reactions. The outcomes revealed that the nano-catalyst could be successively retrieved without any considerable reduction in its performance (Fig. 8).

To explore the leaking of copper, ICP study of the retrieved catalyst was additionally performed. In accord with the acquired outcomes, no considerable reduction was monitored in the Cu quantity. The Cu amount in the new nano-catalyst and the recovered one was 0.94 and 0.91 mmol g^{-1} , respectively, which revealed the amount of leached Cu for this nano-catalyst is very slight. Transmission electron microscopy of the recovered Cu(II)/GQDs/NiFe2O4 catalyst indicated in Fig. 9. From the obtained TEM micrograph obviously exhibited the fresh and used form of catalyst structure approximately similar. Thus, emphasizing no considerable changes happened during the course of reaction. To investigate the heterogeneous character of the catalytic process, the hot filtration examination for the model reaction in attending Cu(II)/GQDs/NiFe₂O₄ was studied. The model reaction was ceased after half the required reaction time and the nano-catalyst was fully isolated with the aid of a magnet and allowed the reaction to progress for further time (30 min). The product generation was not detected as

Table 2 Synthesis of
1,2,3-triazoles 3a-3o using
Cu(II)/GQDs/NiFe ₂ O ₄ as
catalyst ^a

Entry	R	R ¹	Product ^b	Time /min	Yield /% ^c	Melting point /°C	
						Found	Reported
1	Ph	Ph	3a	20	98	96–97	97–98 [37]
2	4-NO ₂ Ph	Ph	3b	15	98	254-255	254 [37]
3	4-BrPh	Ph	3c	20	92	232-233	232 [37]
4	4-CH ₃ Ph	Ph	3d	16	95	174–176	174–176 [37]
5	4-ClPh	Ph	3e	23	95	227-229	228 [37]
6	4-CH ₃ OPh	Ph	3f	15	98	168	168 [37]
7	Ph	CH ₂ OH	3 g	15	91	116–117	116–118 [<mark>38</mark>]
8	4-NO ₂ Ph	CH ₂ OH	3 h	25	93	200-202	201–202 [38]
9	4-BrPh	CH ₂ OH	3i	15	89	134–135	135–137 [<mark>39</mark>]
10	4-CH ₃ Ph	CH ₂ OH	3j	25	88	124-125	124–125 [<mark>40</mark>]
11	4-ClPh	CH ₂ OH	3k	20	94	144–145	144–145 [<mark>41</mark>]
12	4-NO ₂ Ph	CH_2Br	31	28	90	151-153	152–154 [<mark>42</mark>]
13	PhCH ₂	Ph	3m	8	96	129–131	129–131 [40]
14	PhCH ₂	CH ₂ OH	3n	7	92	75–76	75–77 [41]
15	PhCH ₂	CH ₂ Br	30	12	97	123-125	124–126 [42]

^aReaction conditions: azides **1a–1 g** (1.1 mmol), terminal alkynes **2a–2c** (1 mmol), 0.01 g Cu(II)/GQDs/NiFe₂O₄ at 60 °C in H₂O

 $^b \rm Known$ products identified by comparison of their melting points; some selected compounds identified by $^1 \rm H$ NMR and $^{13} \rm C$ NMR

^cIsolated yields





Fig. 8 Reusability of Cu(II)/GQDs/NiFe $_2O_4$ in the model reaction



Fig. 9 Used Cu(II)/GQDs/NiFe₂O₄ TEM image

monitored by TLC which proves the nano-catalyst is heterogeneous in character.

To validate the advantage of the present research, application of Cu(II)/GQDs/NiFe₂O₄ in producing of 1,2,3-triazoles derivatives in comparison with other formerly reported heterogeneous catalysts is presented in Table 3. In accordance with the results, the introduced catalyst indicates more adequate catalytic performance quickly under environmentally friendly and affordable conditions (water).

Conclusions

We have successfully synthesized a copper heterogeneous catalyst on graphene quantum dots/NiFe₂O₄ nanomagnetic particles as a very stable and recoverable nano-catalyst for the green production of 1,2,3-triazoles derivatives rapidly in aqueous media. The proposed reaction, in which the designed Cu(II)/GQDs/NiFe₂O₄ as a recoverable catalyst, consistent with the principles of green chemistry owing to the following properties: no toxicity, great durability, recovery capability, shorter times of reaction, aqueous medium, and excellent products yields. Above all others, the nanocatalyst could be recovered five runs without any failure in its productivity. So, the nanomagnetic heterogeneous catalyst is might be useful in related industries.

Experimental

Citric acid, sodium hydroxide, nickel nitrate, Fe(III) nitrate, and Cu(II) acetate were purchased from Sigma. UV-Vis absorption investigations were conducted by Hach DR 6000 UV-visible spectrophotometer. Fluorescence examinations were done on Jasco FP-6200 spectrofluorophotometer (Hitachi Japan). The phase crystallinity of the prepared nano-catalyst was studied by a diffractometer at wavelength of 1.540 Å (Philips Company). Transmission electron microscopy (TEM) picture was captured by means of Zeiss electron microscope, LEO 912AB (120 kV), Germany. Fourier transform infrared (FT-IR) spectra were recorded on a Bruker model 470 spectrophotometer. Magnetic characteristics were registered on a vibrating sample magnetometer apparatus (VSM, LDJ9600) at environment temperature. EDS spectroscopy study was conducted with 133 eV resolution (model 7353, Oxford Instruments, UK). Copper determination was carried out by inductively coupled plasma (ICP) technique on a Varian VISTA-PRO. NMR spectra were registered in CDCl₃ on a Bruker Advance 300 MHz

Entry	Catalyst	Solvent	Temp. /°C	Time /min	Yield /% ^a	Ref
1	Cu(II)/hydrotalcite	CH ₃ CN	Rt	360	86	[43]
2	AgN(CN) ₂	H ₂ O/EG	Rt	120	95	[44]
3	Cu(PPh ₃) ₂ NO ₃	Solvent-free	Rt	40	96	[45]
4	Nano-Cu/SiO ₂	DMSO	Rt	25	97	[46]
5	Cu/magnetic zeolite	EtOH:H ₂ O	50	120	91	[14]
6	Cu(II)/GQDs/NiFe2O4	H ₂ O	60	20	98	This study

Table 3Comparison of $Cu(II)/GQDs/NiFe_2O_4$ nano-catalystwith other heterogeneouscatalysts in synthesis of1,2,3-triazole**3a**

^aIsolated yield

instrument. The melting points were measured on an Electrothermal Type 9100 device.

Synthesis of NiFe₂O₄ nanoparticles

Egg white (60 cm^3) was added to 40 cm^3 distilled water and was shaken hard to perfectly mix. Subsequently, 2.9081 g of nickel nitrate hexahydrate (10 mmol) and 8.0800 g of Fe(III) nitrate nonahydrate (20 mmol) were dissolved in the above solution and was stirred hard at ordinary temperature for 2 h. Then, whilst the mixture was agitated, it was warmed to 80 °C until dried. The consequent powder was grinded and then heated at 700 °C for 3 h [15].

Synthesis of graphene quantum dots (GQDs)

GQDs were made from thermally decomposed citric acid [30]. Concisely, 0.2 g citric acid (1.0 mmol) was melted and heated at 200° C for 5 min. Then, the subsequent yellowish liquid was added progressively into 20 cm³ of 0.25 M NaOH solution. Thereafter, the GQDs solution was dialyzed in a 1 kDa dialysis bag for 24 h (dialysate was exchanged every 8 h) to eliminate the unreacted chemicals. The produced graphene quantum dot solution was maintained in 4 °C.

Synthesis of GQDs/NiFe₂O₄

NiFe₂O₄ nanoparticles (1 g, 4.23 mmol) were dispersed in 5 cm³ water for 15 min. Afterwards, 20 cm³ of the GQDs suspension was added to the flask comprising the NiFe₂O₄ nanoparticles and the resulting mixture was shaken for 48 h at 60 °C. The produced GQD-modified NiFe₂O₄ was isolated through the use of a magnet and was washed with distilled water (3×20 cm³) and ethanol (3×20 cm³) and finally dried under low pressure [20].

Synthesis of Cu(II)/GQDs/NiFe₂O₄

GQDs/NiFe₂O₄ (0.5 g) was dispersed in 25 cm³ acetone at ordinary temperature for 30 min. Afterwards, 0.018 g copper(II) acetate (0.1 mmol) was added gently to the flask comprising GQDs/NiFe₂O₄ nanocomposite. The consequent mixture was mechanically shaken for 48 h at ambient temperature. Then, the solid was isolated with the aid of a magnet, washed with water (3×25 cm³) and ethanol (3×25 cm³) and finally dried at 60 °C overnight [6].

Synthetic typical manner for 1,2,3-triazoles 3a–3f

Cu(II)/GQDs/NiFe₂O₄ catalyst [0.05 g, 0.047 mmol Cu(II)] was added to a solution comprising aryl azide (1.1 mmol), terminal alkyne (1.0 mmol) in 3 cm³ water under continuous stirring for 10–30 min at 60 °C. The proceed of the

reaction was controlled using TLC (thin layer chromatography), whenever the reaction was completed, the reaction mixture was diluted with ethyl acetate and the nano-catalyst was isolated with the aid of a magnet, washed with acetone and then dried on a night to be ready to react again. The organic layer was dried over anhydrate sodium sulfate and after that, evaporated to separate the solvent. The remaining part was recrystallized in ethanol to provide corresponding triazole derivatives. The products were recognized by their melting points, ¹³C NMR and ¹H NMR spectroscopy. The selected spectral data of some synthesized products have been included in Supplementary Material.

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