Agar: a natural and environmentally-friendly support composed of copper oxide nanoparticles for the green synthesis of 1,2,3-triazoles

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ABSTRACT
A novel, green and cost-effective heterogeneous nanocatalyst was synthesized by supporting copper (I) oxide nanoparticles on magnetic agar (Cu₂O/Agar@Fe₃O₄). Then, it was characterized with multiple techniques, such as scanning electron microscopy and transmission electron microscopy images, energy-dispersive X-ray analysis, Fourier-transform infrared (FT-IR) spectroscopy, thermogravimetric (TG) analysis, X-ray diffraction pattern, vibrating sample magnetometer curve, and inductively coupled plasma analysis. The catalytic activity of the newly designed catalyst was investigated in a one-pot three-component reaction of alkyl halides, sodium azide, and alkynes to obtain 1,4-disubstituted 1,2,3-triazoles in high yields in water–ethanol media. The present catalyst was simply separated from the reaction media by an external magnet and reused at least five subsequent runs without significant activity loss.

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1. Introduction

Recently, the attention of science and technology has turned more into eco-friendly, biocompatible, and sustainable resources. One of the natural, most available and unique compounds in nature is polysaccharides, which are divided into different forms. Agar is a well-known polysaccharide derived from the natural polysaccharide agarose, which forms the supporting structure in the cell walls of certain species of algae (1,2).

The background of agar and agarose dates back to centenaries and these compounds were universally applied following the introduction of the microbiology. The jelly attributes of agar were prime identified by a Chinese Emperor in the 1500s. The agar-production industry was originated from Japan. Today, the manufacturing of agar is beneficial and has created a competitive industry (3).

The two sorts of seaweeds principally used for commercially producing agar are Gracilaria and Gelidium. Gelidium is used for the production of pharmaceutical-grade agar and agarose. Recently different kinds of catalysts, made from synthetic and natural polymers as substrates and metals with noticeable and eye-catching catalytic properties, have been investigated. Since agar contains functional groups which can efficiently coordinate with various transition metals such as Zn, Pt, Pd, Cd, Ni, Co, Mn, and Cu can be chosen as natural support in transition metal-containing catalysts (4–6).
Among these transition metals, copper has newly attracted great attention due to its abundance, cost-effectiveness, and eco-friendliness and its ability to catalyze a wide range of reactions. Copper nanoparticles (NPs) have been extensively used by chemists and physicists due to their specific properties at the nanoscale and the wide assortment of their applications in antimicrobial, magnetic, and electrical devices (7–9).

Metals-based nanoparticles have been widely used as catalysts to perform various organic reactions, such as oxidation and reduction reactions, A³ coupling, cross-coupling, tandem, multicomponent reactions, oxidative coupling, click and cycloaddition reactions because of their suitable properties to promote the reactions under green and sustainable procedure (10–25).

Cu and Cu-based NPs have been extensively employed to catalyze Huisgen 1,3-dipolar cycloaddition reaction, this reaction between azides and terminal alkynes is the most well-known click reaction and has demonstrated a broad range of applications in the industry, synthetic chemistry, and biology, such as anti-HIV activity, antimicrobial activity, anti-cancer, anti-convulsion, anti-allergic, etc. (26–28). Although this reaction has been performed by both homogenous and heterogeneous catalysts, heterogeneous ones are more preferred because of their facile separation from the reaction media without centrifuge and filtration and it can be reused several times (29,30). In the absence of the catalysts, slow rate and low yield have been observed (31). Therefore, in this work, we designed, prepared, and characterized a novel, efficient, and green magnetic heterogeneous nanocatalyst based on agar biomolecule, as shown in Figure 1. In the next step, we studied its catalytic activity in a one-pot three-component reaction of various alkyl halides, sodium azide, and different terminal alkynes under eco-friendly conditions and various

![Figure 1. Preparation of the Cu₂O/Agar/Fe₃O₄ nanocatalyst.](image-url)
triazole derivatives were synthesized with high yields in almost short reaction times (Scheme 1).

2. Experimental

2.1. General methods

Agar and other chemicals, solvents, and reagents were purchased from Merck Company. Melting points (MPs) were measured on an electrothermal 9100 apparatus. Fourier-transform infrared (FT-IR) spectra were recorded on a Shimadzu IR-470 spectrometer by KBr pellets. 1H nuclear magnetic resonance (NMR) spectra were recorded in CDCl 3 solvent with a Bruker DRX-500 Avance spectrometer at 500 MHz. Scanning electron microscopy (SEM) images were obtained on a MRIA3 TESCAN device. Transmission electron microscopy (TEM) was provided on a Philips CM200 in an acceleration voltage of 200 kV. Energy-dispersive X-ray (EDX) analysis was recorded on Numerix DXP-X10P spectrometer. Analytical thin layer chromatography (TLC) was performed on pre-coated silica gel 60F254 plates. MPs were obtained in capillary tubes by Electrothermal 9100 instrument. Thermogravimetric analysis (TGA) was performed using a Bahr-STA 504 instrument at a heating rate of 10 °C min\(^{-1}\) in air atmosphere. X-ray diffraction (XRD) measurements were carried out using a JEOL JDX-8030 (30 kV, 20 mA). Vibrating sample magnetometer (VSM) was carried out by LBKFB model-magnetic Danhigh Kavir Company. Inductively coupled plasma (ICP) analysis was provided on a Shimadzu ICPS-7000.

2.2. Synthesis of Cu\(_2\)O/Agar@Fe\(_3\)O\(_4\) nanocatalyst

Powder agar (1.00 g) was added to 20 mL of deionized water and was stirred at 70°C until it was dissolved properly. Then FeCl\(_2\)·4H\(_2\)O (0.79 g) and FeCl\(_3\)·6H\(_2\)O (2.16 g) with a molar ratio of (1:2) were added to the mixture and N\(_2\) atmosphere was applied, after a while, when the temperature of the mixture increased to 70°C, 5 mL of (25\%wt) ammonia solution was gradually added to the mixture until a black solution (Agar@Fe\(_3\)O\(_4\)) was obtained. Then, the temperature of the mixture was cooled down to room temperature and CuCl\(_2\)·2H\(_2\)O (0.09 g) dissolved in 10 mL of deionized water was added to the mixture; afterwards, the temperature of the mixture was increased to 70°C anew and stirred for 24 h, in the first 3 h of stirring, ascorbic acid (.044 g) was added to mixture gradually in order to reduce copper oxide (Cu\(^{2+}\)) to (Cu\(^{+}\)) (8). After the completion of the reaction, the magnetic agar supported with copper(I) oxide was magnetically separated and washed with deionized water and ethanol several times, finally, the resultant precipitate was dried at 90°C to obtain a described nanocatalyst.

2.4. General procedure for the synthesis of 1,2,3-triazole

Alkyl halide (1 mmol), alkyn (1 mmol), and sodium azide (1 mmol) were dissolved in the mixture of (2.5 mL) water and (2.5 mL) ethanol in the presence of Cu\(_2\)O/Agar@Fe\(_3\)O\(_4\) nanostructure (25 mg) and the mixture was stirred at 70°C for 2.5 h (Table 1). After the completion of the reaction (monitored by TLC), the mixture was cooled to room temperature and the catalyst was separated by using an external magnet and washed with deionized water and ethanol several times. All the products were confirmed by the comparison of their MPs with the authentic samples mentioned in the literature as well as their characterized NMR spectra.

2.5. Characterization data (1\(^1\)H NMR spectra) of the products

1-Benzyl-4-phenyl-1H-1,2,3-triazole
1H NMR (500 MHz, CDCl3): δ: 5.60 s (2H, CH2), 7.33–7.35 m (3H, H aromatic), 7.4–7.44 m (5H, H aromatic), 7.68 s (1H, 5-H), 7.81–7.83 d (2H, H aromatic).

1-(4-Nitrobenzyl)-4-phenyl-1H-1,2,3-triazole
1H NMR (300 MHz, CDCl3): δ = 5.69 (s, 2H), 7.30–7.45 (m, 5H), 7.75–7.81 (m, 3H), 8.18–8.20 (m, 2H).

1-(4-Bromobenzyl)-4-phenyl-1H-1,2,3-triazole
1H NMR (300 MHz, CDCl3): δ = 5.65 (s, 2H), 7.32–7.43 (m, 5H), 7.64 (d, 2H), 7.89 (d, 2H), 8.63 (s, 1H).

1-(2-Chlorobenzyl)-4-phenyl-1H-1,2,3-triazole
1H NMR (300 MHz, CDCl3): δ = 5.71 (s, 2H), 7.22–7.36 (m, 5H), 7.47–7.49 (m, 3H), 8.14–8.16 (d, 2H).

1-(4-Methylbenzyl)-4-phenyl-1H-1,2,3-triazole
1H NMR (300 MHz, CDCl3): δ = 2.35 (s, 3H), 5.51 (s, 2H), 7.18–7.20 (m, 4H), 7.65 (s, 1H), 7.77–7.81 (m, 2H).

(1-Benzyl-1H-1,2,3-triazole-4-yl)methanol
1H NMR (300 MHz, CDCl3): δ = 3.29 (br s, 1H), 4.73 (s, 2H), 5.47 (s, 2H), 7.35–7.41 (m, 5H), 7.47 (s, 1H).

3. Results and discussion

3.1. Characterization of Cu2O/Agar@Fe3O4 nanocatalyst

The synthetic steps of Cu2O/Agar@Fe3O4 nanocatalyst from Gelidium seaweed is shown in Figure 1. Then, it was characterized by various conventional techniques. Initially, FT-IR of agar and Cu2O/Agar@Fe3O4 nanocatalyst were provided (Figure 2). In the FT-IR spectrum of Cu2O/Agar@Fe3O4 nanocatalyst, the characteristic adsorption band of Fe–O and Cu–O was observed at around 561–631 cm⁻¹ that corresponds to the stretching vibrations and the peak at 1607 cm⁻¹ is ascribed to the stretching vibration of C=C which is present in the agar molecules. The broadband at 3300–3500 cm⁻¹ is due to –OH stretching vibrations. The decrease in the intensity of the peak related to OH stretching vibration can be a reliable proof to confirm the immobilization of Cu2O and Fe3O4 nanoparticles on agar substrate.

SEM image analysis was used to study the surface morphology and size distribution of the Cu2O/Agar@Fe3O4 nanocatalyst. As can be seen in Figure 3, spherical nanoparticles were dispersed properly on the agar surface. The size range of most particles was 11 up to 19 nm. Spherical morphology of the nanoparticles may improve the catalytic efficiency by extending the surface area and increasing available catalytic sites.

Figure 4 shows that the TEM image of Cu2O/Agar@Fe3O4 nanostructure was provided to confirm core@shell morphology of the nanocatalyst, which has shown that the Fe3O4 nanoparticles are embedded properly into the agar matrix.

EDX analysis was used to detect the chemical composition of the prepared nanocatalyst. As illustrated in Figure 5, the peaks, which have appeared in the EDX pattern of the nanocatalyst, clearly confirm the existence of oxygen, iron, and copper elements in the nanostructure.

As illustrated in Figure 6, TGA was performed in the range of 50–800°C under atmospheric conditions. The curve of agar (a) shows the first weight loss at around 80°C due to the evaporation of adsorbed water and the total weight loss is around 700°C, about 98%, due to the decomposition of agar. The Cu2O/Agar@Fe3O4 nanocatalyst (b) lost its weight about 10% when the temperature was above 310°C because of the evaporation of adsorbed water. The residual weight is due to the presence of Cu2O and Fe3O4 and agar. A comparison
between Figure 6(a) and Figure 6(b) shows two points. The first one, at a temperature of 100–270°C, the composite can hold up a higher amount of water than pure agar. The second one, the agar at above 275°C starts to decompose and at around 700°C it is completely decomposed, but the composite lost its weight at the same temperature range with very little slope, and this represents a very high thermal resistance of the composite.

The XRD diffraction analysis was used to determine the crystalline or amorphous nature of Cu₂O/Agar@Fe₃O₄ nanocatalyst shown in Figure 7. The XRD pattern shows five intense and sharp peaks at 2θ values 30.32°, 35.81°, 42.95°, 57.78°, and 62.90° corresponding to Fe₃O₄ nanoparticles (JCPDS no. 85-1436) and three overlapped peaks at 2θ = 29.85°, 36.02°, and 43.16° relating to Cu₂O nanoparticles (JCPDS no. 78-2076). The XRD pattern also shows no peak related to agar due to the amorphous nature of agar. All peaks are in a good

Figure 3. SEM images of Cu₂O/Agar@Fe₃O₄ nanocatalyst in different magnifications.

Figure 4. TEM image of Cu₂O/Agar@Fe₃O₄ nanocatalyst.
agreement with standard XRD patterns. Furthermore, by the Scherer equation, the average crystal size was obtained \( d = 15.4 \text{ nm} \) which is matched to the size of the particles of the SEM images.

The magnetic property of Cu2O/Agar@Fe3O4 nanostructure was investigated by VSM at room temperature. As it shown in Figure 8, the value of the saturation magnetization was 56.3 emu g\(^{-1}\). This indicates the proper amount of Fe3O4 NPs loaded in the catalyst. Because of the high magnetic potential of the catalyst, it can be easily separated from reaction media by an external magnet.

3.2. Application of nanocatalyst in organic synthesis

The catalytic activity of the prepared nanostructure was investigated in the multicomponent cycloaddition reaction of organic azides (which are obtained from the corresponding sodium azide and halides) and terminal alkynes. The reaction of phenyl acetylene, benzyl bromide, and sodium azide were chosen as the model reaction and the effect of different experimental conditions on the yield of the reaction was perused. The phenyl acetylene, benzyl bromide, and sodium azide were added to a suspension of Cu2O/Agar@Fe3O4 NPs as a heterogeneous catalyst (25 mg) in a mixture of water/ethanol solvents. The reaction mixture was stirred for 2.5 h at 70°C, and was cooled to room temperature and the catalyst was easily removed using an external magnet and washed several times with chloroform to separate the adsorbed organic compounds. Eventually, the solvent was eliminated under vacuum to afford the pure 1,4-disubstituted 1,2,3-triazole in 95% yield. As can be seen in Table 1 (entry 1), while the model reaction was done in the absence of Cu2O/Agar@Fe3O4 NPs catalyst, after 2.5 h the desired product was not formed.

Choosing a suitable solvent is a very important step in order to design a green and environment-friendly, safe and low-cost reaction procedure. The principle of green chemistry expresses the goal to minimize the environmental impact resulting from the utilization of toxic and hazardous solvents in the chemical
production (32–35). In this work, all the solvents used in optimization are safe and environmentally benign. To choose the appropriate medium of the reaction, the model reaction was tested in diverse green solvents and solvent–cosolvent mixtures: water, ethanol, acetone, and water–ethanol. Solvent-free mechanochemical ball milling was tested and the results are summarized in Table 1. Performing the reaction in water–ethanol as a solvent-cosolvent mixture gave the foremost result (Table 1, Entry 4).

After optimization of the reaction conditions in order to generalize the optimum conditions and to show the versatility of this procedure, different derivatives of terminal alkynes and benzyl halides were used. As expected, the high yields of triazole were obtained. The results are summarized in Table 2.

The proposed reaction mechanism is illustrated in Scheme 2. As depicted in the scheme, the organic azide is formed through a nucleophilic attack of azide ion to alkyl bromide and releasing Br\(^-\) as a leaving group. The Cu species reacts with a terminal alkyne to form copper acetylide. The 1,3-dipolar cyclization of the resulting copper acetylide, an organic azide, and the subsequent protonation leads to the formation of 1,2,3-
Table 2. Huisgen 1,3-dipolar cycloadditions catalyzed by Cu$_2$O/Agar@Fe$_3$O$_4$ nanocatalyst.

\[
\text{R}^1\text{X} + \text{NaN}_3 + \text{R}^2 \xrightarrow{\text{Cu}_2\text{O}/\text{Agar@Fe}_3\text{O}_4 \text{ nanocatalyst} \text{\text{water-ethanol}} \text{}} \text{Product}
\]

| Entry | R$^1$       | X    | R$^2$     | Product | Time (h) | Yield$^b$ (%) | MP (°C)          |
|-------|-------------|------|-----------|---------|----------|---------------|-----------------|
| 1     | PhCH$_2$    | Br   | Ph        | ![Product Image](ProductImage1.png) | 2.5      | 95            | 128–130 (129–130) (36) |
| 2     | 4-NO$_2$C$_6$H$_4$CH$_2$ | Br | Ph        | ![Product Image](ProductImage2.png) | 4        | 81            | 150–153 (150–155) (36) |
| 3     | 4-BrC$_6$H$_4$CH$_2$ | Br | Ph        | ![Product Image](ProductImage3.png) | 2        | 97            | 152–154 (152–152.5) (37) |
| 4     | 2-ClC$_6$H$_4$CH$_2$ | Cl | Ph        | ![Product Image](ProductImage4.png) | 3.5      | 86            | 78–81 (79–81) (36) |
| 5     | 4-CH$_3$C$_6$H$_4$CH$_2$ | Br | Ph        | ![Product Image](ProductImage5.png) | 4        | 81            | 106–108 (104–106) (38) |
| 6     | PhCH$_2$    | Br   | HOCH$_2$  | ![Product Image](ProductImage6.png) | 5        | 91            | 77–78 (77–79) (38) |
| No. | Halogen | Substituent | Yield % | Reaction Temperature |
|-----|---------|-------------|---------|----------------------|
| 7   | 4-NO$_2$C$_6$H$_4$CH$_2$ Br | HOCH$_2$ | 80      | 130–132              |
| 8   | 4-BrC$_6$H$_4$CH$_2$ Br | HOCH$_2$ | 92      | 116–119              |
| 9   | 2-ClC$_6$H$_4$CH$_2$ Cl | HOCH$_2$ | 81      | 105–108 (110–112) (39) |
| 10  | 4-CH$_3$C$_6$H$_4$CH$_2$ Br | HOCH$_2$ | 80      | 89–92 (91.5–92.5) (40) |

*Reaction conditions: primary halide (1 mmol), NaN$_3$ (1 mmol), alkyne (1 mmol), Cu$_2$O/Agar@Fe$_3$O$_4$ (25 mg), water–ethanol (5 mL), 70°C.*

*Isolated yield.*
triazole and the regeneration of Cu$_2$O/Agar@Fe$_3$O$_4$ nanocatalyst (41).

To determine catalyst reusability, the model reaction was carried out using Cu$_2$O/Agar@Fe$_3$O$_4$ nanocatalyst to synthesize the desired product. Because of its magnetic property, the synthesized catalyst could be easily separated from the reaction media and after washing with the appropriate solvents, it can be dried and re-used. As can be observed in Figure 9, the catalyst can be reused at least five times without any significant decrease in the reaction yield. This observation indicates that no considerable change in the Cu loading after five runs, which was exactly supported by the ICP analysis. The ICP analysis shows only 6% of Cu leached into the reaction media after five consecutive runs.

4. Conclusions

We introduce a new, green, and magnetically separable heterogeneous Cu$_2$O polymer-based catalyst during a very facile and simple preparation method using safe, abundant, and cost-effective raw materials. This naturally originated nanocatalyst was then employed in a one-pot
three-component reaction of sodium azide, phenyl acetylene, and benzyl bromide to obtain 1,2,3-triazole derivatives, which are present in lots of biologically active compounds. Due to the high efficiency of the catalyst, applying harsh reaction condition, such as performing the reaction at extremely high temperatures or under reflux condition, was not necessary. All the identification techniques, such as FT-IR, SEM, TEM, EDX, TGA, XRD, ICP, and VSM analyses, confirmed the formation of as-described catalyst. According to the mentioned properties, and following the principle of green chemistry, this catalyst can be used in the industry and in laboratories. The best advantages of this catalyst include facile preparation, simple separation, and providing a safe reaction pathway to produce a variety of 1,2,3-triazole derivatives with high atom efficiency (Table 2). The organic products were confirmed via their MPs and 1H NMR. Moreover, the SEM and TEM images show the proper distribution with the average range of most particles about 15 nm (Figures 3 and 4). The EDX analysis showed the existence of carbon, oxygen, iron, and copper elements in Cu2O/Agar@Fe3O4NPs (Figure 5). The very high thermal resistance of the nanocatalyst was confirmed by the TGA.

Disclosure statement
No potential conflict of interest was reported by the authors.

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