Green Synthesis of New Category of Pyrano[3,2-c]Chromene-Diones Catalyzed by Nanocomposite as Fe3O4@SiO2-Propyl Covalented Dapsone-Copper Complex

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Nanomagnetic dapsone-Cu supported on the silica-coated Fe3O4 (Fe3O4@SiO2-pr@dapsone-Cu) nanocomposite was synthesized and characterized by Fourier transform infrared (FT-IR), energy-dispersive X-ray (EDX), X-ray diffraction (XRD), field emission scanning electron microscope (FE-SEM), transmission electron microscopy (TEM), zeta potential, vibrating sample magnetometer (VSM), and thermogravimetric analysis (TGA). This newly synthesized nanocomposite was chosen to act as a green, efficient, and recyclable Lewis acid for the multicomponent synthesis of new derivatives of pyrano[3,2-c] chromene-diones through the reaction of aromatic aldehydes, indandione, and 4-hydroxycoumarin in water. All of the synthesized compounds are new and are recognized by FT-IR, NMR, and elemental analysis; this avenue is new and has advantages such as short reaction times, high productivity, economical synthesis, and use of green solvent, H2O, as a medium. The catalyst is magnetically recoverable and can be used after six runs without a decrease in the efficiency.

Keywords: dapsone-cu, silica-coated Fe3O4, silica, pyranochromenes, nanoorganometallic chemistry

INTRODUCTION

Multicomponent reactions (MCRs) are useful avenue for the synthesis of organic compounds. This reaction is a combination of at least three components in a one-pot domino (Li and Chan, 1997; Grieco, 1998; Dömling and Uzi, 2000; Dömling, 2002; Hosseini-Zare et al., 2012). MCRs have benefits such as higher atom economy and selectivity and production of complex molecules with low by-products. Nowadays, MCRs have attracted a lot of interest in organic transformation (Bienaymé et al., 2000; Kandhasamy and Gnanasambandam, 2009; Müller 2014).

Pyranochromenes have various biological activities such as antifungal (Ouf et al., 2014), antibacterial (Zhang et al., 2012), antihyperglycaemic and antidysslipidemic (Kumar et al., 2009), anticancer (Lee et al., 2009), cytotoxic (Magiatis et al., 1998), anti-HIV (Kongkathip et al., 2005), anti-HBV (Su et al., 2009), antiviral (Barnard et al., 2002), antiproliferative (Mao et al., 2019), anti-inflammatory (Symeonidis et al., 2009), antinociceptive (Lima et al., 2006), and antituberculosis (Xu et al., 2006). They are also synthetic intermediates for alkaloids, drug candidates, and clinical pharmaceuticals (Williams and Cox, 2003).

Magnetic nanoparticles (MNPs) as catalysts are interesting materials because of their high surface area, unique magnetic properties, and high catalytic activity. They were used as magnetic fluids...
catalysis and magnetic resonance imaging (MRI) data storage and environmental remediation (Rezaee Nezhad et al., 2014; Mokhtary, 2016; Rezayati et al., 2016; Fekri and Maleki, 2017; Abbasi et al., 2017; Fekri et al., 2020; Sardarian et al., 2019; Inaloo et al., 2020a; 2020b; 2020c; 2020d).

To the best of our knowledge, there are no reports on the use of Fe3O4@SiO2-propyl-loaded dapsone-copper as a catalyst for the synthesis of pyranochromene-diones via multicomponent reactions of aldehydes, indandione, and 4-hydroxycoumarin results.

**EXPERIMENTAL**

**Material and Method**

The X-ray diffraction (XRD), transmission electron microscopy (TEM), scanning electron microscopy (SEM), thermogravimetric analysis (TGA), and zeta potential analysis for synthesized MNPs were analyzed on X-PRTPRO (Netherlands) XRD, TEM Jeol model 3,010, Philips XL 30 scanning electron microscope (SEM, Netherlands), Q600 (made in America), and ZEN3600 (England) instrument, respectively. Fourier transform infrared (FT-IR) spectra of organic compounds were carried out by Shimadzu 8400 spectrophotometer (FT-IR-470, Japan). 1H and 13C NMR spectra were determined using a Bruker DRX Avance instrument at 500 or 300 MHz.

**General Procedure for the Synthesis of Fe3O4@SiO2@dapsone-Cu MNPs**

A mixture of aldehyde (1.0 mmol), indan-1,3-dione (2.0 mmol), 4-hydroxycoumarin (1 mmol), and 0.05 g Fe3O4@SiO2@dapsone-Cu MNPs was stirred at room temperature in 10 ml distilled water for the required reaction time as indicated by TLC (TLC silica gel 60 F250, ethyl acetate : n-hexane 1 : 4). After completion of the reaction, the resulting mixture was filtered in the presence of an efficient magnetic bar to separate the catalyst. The catalyst was washed with 10 ml ethanol and reused. The crude products were collected and dried.

**Characterization Data**

7-(4-Nitrophenyl)-6H-indeno [2′,1′:5,6]pyrano[3,2-c]chromene-6,8(7H)-dione 4a: m. p. 203–205°C, FT-IR (KBr, cm⁻¹): 1739 (C=O str), 1701 (C=O str), 1,561 (Asymmetric NO2 str or aromatic C=C str), 1,510, 1,456 (aromatic C-C str), 1,348 (symmetric NO2 str), 1,257 (C-O str), 1,099. H NMR (DMSO-d6, 400 MHz): δH 5.31 (s, 1H, chiral C-H), 6.84–6.88 (m, 1H, Ar), 7.00–7.04 (m, 1H, Ar), 7.18 (d, J = 8.0Hz, 1H, Ar), 7.23 (d, J = 2.4Hz, 1H, Ar), 7.31 (d, J = 8.0Hz, 1H, Ar), 7.35 (s, 1H, Ar), 7.55 (d, J = 8.0Hz, 2H, Ar), 7.85–7.95 (m, 2H, Ar), 8.06–8.14 (m, 1H, Ar) ppm. 13C NMR (DMSO-d6, 100 MHz): δC 57.60, 123.98 (Ar), 123.33 (Ar), 137.89 (Ar), 133.98 (Ar), 139.09 (two peaks, Ar), 141.23 (Ar), 145.81 (Ar), 149.87 (Ar), 198.76 (C=O) ppm. 201.17 (C=O) ppm. Anal. Calcd. for C25H13NO6: C, 70.93; H, 3.31. Found: C, 70.93; H, 3.30; N, 3.32.

7-(3-Nitrophenyl)-6H-indeno [2′,1′:5,6]pyrano[3,2-c]chromene-6,8(7H)-dione 4b: m. p. 176–177°C, FT-IR (KBr, cm⁻¹): 1736 (C=O str), 1700 (C=O str), 1,660, 1,580 (asymmetric NO2 or aromatic C=C str), 1,430 (aromatic C=C str), 1,330 (symmetric NO2 str), 1,275 (C-O str). 1H NMR (DMSO-d6, 100 MHz): δH 5.33 (s, 1H, chiral C-H), 6.85 (t, J = 7.2Hz, 1H, Ar), 7.00 (t, J = 7.2Hz, 1H, Ar), 7.22 (d, J = 8.4Hz, 2H, Ar), 7.31 (d, J = 8.4Hz, 1H, Ar), 7.35 (d, J = 2.0Hz, 1H, Ar), 7.52 (t, J = 8.0Hz, 1H, Ar), 7.78 (d, J = 8.0Hz, 1H, Ar), 7.85–7.94 (m, 2H, Ar), 8.00–8.02 (m, 1H, Ar), 8.18 (d, J = 2.0Hz, 1H, Ar) ppm. 13C NMR (DMSO-d6, 100 MHz): δC 58.30 (chiral C), 112.00 (two peaks, Ar), 113.54 (Ar), 114.00 (Ar), 118.96 (Ar), 119.18 (Ar), 121.80 (Ar), 122.03 (Ar), 123.25 (Ar), 123.33 (Ar), 124.76 (Ar), 126.71 (two peaks, Ar), 131.06 (Ar), 131.21 (Ar), 132.33 (Ar), 137.89 (Ar), 133.98 (Ar), 139.09 (two peaks, Ar), 141.23 (Ar), 145.81 (Ar), 149.87 (Ar), 197.86 (C=O), 201.17 (C=O) ppm. Anal. Calcd. for C25H13NO6: C, 70.92; H, 3.09; N, 3.31. Found: C, 70.95; H, 3.07; N, 3.36.

7-(4-Bromophenyl)-6H-indeno [2′,1′:5,6]pyrano[3,2-c]chromene-6,8(7H)-dione 4c: m. p. 276–278°C, FT-IR (KBr, cm⁻¹): 1735 (C=O str), 1700 (C=O str), 1,650, 1,457 (aromatic C=C str), 1,215 (C-O str). 1H NMR (DMSO-d6, 400 MHz): δH 5.13 (s, 1H, chiral C-H), 6.85 (t, J = 7.2Hz, 1H, Ar), 7.01 (t, J = 7.2Hz, 1H, Ar), 7.13 (d, J = 8.0Hz, 1H, Ar), 7.17 (d, J = 8.4Hz, 2H, Ar), 7.31 (dd, J = 3.2Hz, J = 5.6Hz, 2H, Ar), 7.35 (dd, J = 1.6Hz, J = 6.4Hz, 2H, Ar), 7.84–7.93 (m, 2H, Ar) ppm. 13C NMR (DMSO-d6, 100 MHz): δC 58.48 (chiral C), 111.91 (Ar), 114.32 (Ar), 118.95 (Ar), 119.00 (Ar), 119.99 (Ar), 121.60 (Ar), 123.25 (Ar), 124.76 (Ar), 126.71 (two peaks, Ar), 131.06 (Ar), 131.21 (Ar), 131.27 (Ar), 136.44 (Ar), 136.58 (Ar), 141.35 (Ar), 142.38 (Ar), 142.61 (Ar), 144.32 (Ar), 197.56 (C=O), 200.26.

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(=O) ppm. Anal. Calcd. for C_{25}H_{13}BrO_{4}: C, 65.66; H, 2.87. Found: C, 65.65; H, 3.07; N, 2.86.

7-(3-Hydroxyphenyl)-6H-indeno [2',1':5,6]pyrano [3,2-c] chromene-6,8(7H)-dione 4f: m. p. 289–290°C, FT-IR (KBr, cm⁻¹): 1737 (C=O str), 1705 (C=O str), 1,650, 1,608 (aromatic C=C str), 1,538, 1,224 (C-O str). ¹H NMR (DMSO-d₆, 400 MHz): δ_{H} 5.21 (s, 1H, chiral C-H), 6.78 (t, J = 7.8Hz, 1H, Ar), 7.02 (t, J = 7.5Hz, 1H, Ar), 7.18–7.22 (m, 2H, Ar), 7.30 (t, J = 8.5Hz, 3H, Ar), 7.65 (d, J = 8.6Hz, 1H, Ar), 7.87–7.94 (m, 3H, Ar), 8.31 (d, J = 7.8Hz, 1H, Ar) ppm. ¹³C NMR (DMSO-d₆, 100 MHz): δ_{C} 56.78 (chiral C), 113.09 (two peaks, Ar), 114.56 (Ar), 116.12 (Ar), 116.91 (Ar), 116.99 (Ar), 118.13 (Ar), 118.76 (Ar), 121.43 (two peaks, Ar), 121.26 (two peaks, Ar), 123.54 (Ar), 125.57 (Ar), 126.47 (Ar), 128.46 (Ar), 134.09 (Ar), 138.11 (two peaks, Ar), 147.87 (Ar), 155.08 (Ar), 197.34 (C=O), 200.07 (C=O) ppm. Anal. Calcd. for C_{25}H_{13}NO_{4}: C, 75.98; H, 3.45; N, 3.69. Found: C, 75.93; H, 3.43; N, 3.65.

7-Phenyl-6H-indeno [2',1':5,6]pyrano [3,2-c] chromene-6,8(7H)-dione 4g: m. p. 203–205 °C, FT-IR (KBr, cm⁻¹): 3,098 (aromatic C-H str), 1735 (C=O str), 1702 (C=O str), 1,603 (aromatic C=C str), 1,408, 1,365, 1,231, 1,211 (C-O str). ¹H NMR (DMSO-d₆, 400 MHz): δ_{H} 5.10 (s, 1H, chiral C-H), 6.35–6.56 (m, 3H, Ar), 6.68 (d, J = 7.8Hz, 2H, Ar), 6.86 (s, 1H, Ar), 6.98–7.11 (m, 2H, Ar), 7.28 (d, J = 6.8Hz, 3H, Ar), 7.34 (d, J = 8.2Hz, 2H, Ar) ppm. ¹³C NMR (DMSO-d₆, 100 MHz): δ_{C} 58.52 (chiral C), 114.23 (Ar), 114.38 (Ar), 116.01 (Ar), 116.98 (Ar), 117.45 (Ar), 117.67 (Ar), 117.89 (Ar), 121.32 (Ar), 121.67 (Ar), 123.55 (Ar), 124.78 (Ar), 126.98 (Ar), 127.46 (Ar), 129.00 (Ar), 132.65 (two peaks, Ar), 136.89 (Ar), 138.91 (Ar), 141.24 (Ar), 143.67 (Ar), 196.34 (C=O), 200.32 (C=O) ppm. Anal. Calcd. for C_{26}H_{14}O_{4}: C, 79.36; H, 3.73. Found: C, 79.31; H, 3.77.

7-(p-Tolyl)-6H-indeno [2',1':5,6]pyrano [3,2-c] chromene-6,8(7H)-dione 4h: m. p. 234–235°C, FT-IR (KBr, cm⁻¹): 1734 (C=O str), 1701 (C=O str), 1,576 (aromatic C=C str), 1,435 (aromatic C=C str), 1,387, 1,234 (C-O str), 1,114 (H NMR (DMSO-d₆, 400 MHz): δ_{H} 2.35 (s, 3H, Ph-CH₃), 5.31 (s, 1H, chiral C-H), 6.87 (d, J = 7.8Hz, 2H, Ar), 6.87–7.09 (m, 2H, Ar), 7.23 (J = 8.5Hz, 1H, Ar), 7.56 (d, J = 7.8Hz, 2H, Ar), 7.67–7.83 (m, 3H, Ar), 8.01–8.09 (m, 2H, Ar) ppm. ¹³C NMR (DMSO-d₆, 100 MHz): δ_{C} 34.23 (benzylic carbon), 58.65 (chiral C), 112.67 (Ar), 114.00 (Ar), 115.34 (Ar), 116.46 (Ar), 118.87 (two peaks, Ar), 119.45 (Ar), 121.33 (Ar), 125.67 (Ar), 126.23 (Ar), 128.98
FIGURE 1 | (A) FE-SEM and (B) TEM images of synthesized Fe₃O₄@SiO₂-pr@dapsone-Cu.

FIGURE 2 | FT-IR spectra of Fe₃O₄@SiO₂-propyl@dapsone-Cu MNPs.
RESULT AND DISCUSSION

Synthesis and Characterization

In order to prepare nanocatalyst, initially, Fe3O4 MNPs were modified with silica and then with chloropropyl silane via chemical bonds to obtain Fe3O4@SiO2-pr. In the next step, Fe3O4@SiO2-propyl was covalently by substitution reaction with dapsone to prepare Fe3O4@SiO2-propyl loaded dapsone.

This nanocatalyst was treated with copper chloride to produce Fe3O4@SiO2-propyl@dapsone-Cu (Scheme 1). The structure of the prepared nanocatalyst was studied and fully characterized using FT-IR, energy-dispersive X-ray (EDX), XRD, zeta potential, TEM, and field emission scanning electron microscope (FE-SEM) analysis.

As shown in Figure 1 (FE-SEM and TEM), the magnetic nanoparticles have a spherical shape with an average diameter of 14–38 nm. The synthesized nanoparticles have aggregated well.

Figure 2 shows FT-IR spectra of Fe3O4@SiO2-propyl@dapsone-Cu MNPs. Wavenumbers of Fe-O bonds of Fe3O4 appear in 569 and 467 cm⁻¹ in Fe3O4@SiO2-propyl@dapsone-Cu MNPs. The peaks positioned at 2,928 cm⁻¹ are assigned to...
aliphatic C-H bonds of Fe₃O₄@SiO₂-propyl@dapsone-Cu MNPs. The peak at 1,102 cm⁻¹ and 1,141 cm⁻¹ is attributed to Si-O-Si stretching modes of Fe₃O₄@SiO₂-propyl@dapsone-Cu.

Further, the other peaks were seen as 1,594 (S=O stretching), 1,628 (aromatic C=C stretching), 3,371 (N-H stretching), and 1,343 cm⁻¹ (C-N stretching) in Fe₃O₄@SiO₂-propyl@dapsone-Cu.

The XRD pattern of synthesized nanoparticles showed the sharp diffraction peaks with 2θ at 18° (111), 30.4° (220), 35.7° (311), 43.3° (400), 53.6° (422), 57.7° (511), 63.0° (440), and 74.6° (533) (Figure 3A), which indicate that the MNPs have highly crystalline cubic spinel structure of the magnetite and matched with the diffraction patterns of the standard Fe₃O₄ (JCPD 19–0629). This confirmed the stability of the crystalline phase of the magnetite core in the structure after silica coating, condensation, and complexation process. The absence of an amorphous peak in pattern confirmed the crystalline structure. Using Debye–Scherrer equation, the mean size of crystallite was calculated as 12.1 nm from the XRD pattern (crystallite shape factor: 0.9 and λ CuKα = 1.54060 Å). This value is lower than the size obtained by FE-SEM and TEM due to the fact that some crystallite forms a particle. Also, the d-spacing and full width at half maximum

**FIGURE 4** | (A) Zeta potential and (B) EDX analysis of Fe₃O₄@SiO₂-propyl@dapsone-Cu complex.

**FIGURE 5** | VSM analysis of nanoparticles.
The FWHM of the highest XRD peak (2θ = 35.71°) were obtained as 2.514 Å and 0.6888 (2θ°).

Figure 3B revealed the TGA analysis of synthesized nanoparticles. Two weight losses are observed. The first decrease is related to a temperature below 333°C because of desorption of water and the second weight-loss step at 524°C is due to decomposition of organic compound as dapsone. As shown in Figure 4A, the zeta potential was scanned. The large zeta potential obtained revealed a more stable dispersion of synthesized MNPs. The zeta potential value of dispersed synthesized in deionized water in absence of any electrolyte was +25.1 mV.

The presence of iron, oxygen, nitrogen, carbon, silica, sulfur, and copper, in EDX, revealed the successful synthesis of these nanoparticles.

The magnetic properties of synthesized nanoparticles are shown in Figure 5. The results approve the superparamagnetism behavior.

**Catalytic Application**

In continuation of our study to synthesize pharmaceutical compounds (Fekri et al., 2018a, b; Fekri and Fard, 2016; Zare et al., 2010; Nikpassand et al., 2012; Fekri and Nikpassand 2014), we triggered to use Fe3O4@SiO2-propyl@dapsone-Cu nanoparticles, for the multicomponent synthesis of novel derivatives of pyranochromene-diones via the multicomponent reaction between various aldehydes, indan-1,3-dione, and 4-hydroxycoumarine (Scheme 2).

To complete our assessment, we checked the effect of different conditions in the sample reaction. For example, we treated 4-nitrobenzaldehyde, indandione, and 4-hydroxycoumarin under stirring at room temperature and refluxing in EtOH. The satisfactory results were obtained via the reaction of 4-nitrobenzaldehyde, indandione, and 4-hydroxycoumarin in the presence of 0.05 g of Fe3O4@SiO2-propyl@dapsone-Cu in aqueous media under stirring (Table 1).

To expand the generality and efficiency of this avenue, some aldehydes with electron-donating or electron-withdrawing substituents were treated with indan-1,3-dione and 4-hydroxycoumarin. The results are summarized in Table 2.

As a proposed mechanistic pathway, initially, aldehyde was activated by the nanocatalyst, followed by nucleophilic attack of C-H acid of indan-1,3-diones, together with the departure of water, and chalcone was produced. Nucleophilic attack of 4-hydroxycoumarin to chalcone as Michael addition and then intramolecular cyclization followed by elimination of water lead to product 4 (Scheme 3).

Furthermore, the magnetic nanoparticles are magnetically recoverable and can be reused for six runs.

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**SCHEME 2 | Multicomponent synthesis of pyranochromene-diones.**

**TABLE 1 | The optimization reaction conditions for the synthesis of 4a.**

| Catalyst                  | Condition               | Time (h) | Yield (%) |
|---------------------------|-------------------------|----------|-----------|
| —                         | Refluxing in EtOH       | 48       | 11        |
| HCl (concentrated)        | Refluxing in EtOH, 10drops | 48     | 28        |
| AcOH                      | Refluxing in EtOH, 0.1 ml | 48       | 35        |
| K10                       | Refluxing in EtOH, 0.1 g | 24       | 45        |
| HY-zelite                 | Refluxing in EtOH, 0.1 g | 24       | 51        |
| DBU-Ac                    | Heating at 90°C, 0.5 ml | 30       | 43        |
| DABCO-dAc                 | Heating at 90°C, 0.5 ml | 26       | 47        |
| Nano-Fe3O4                | Refluxing in EtOH, 0.1 g | 12       | 58        |
| Fe3O4@SiO2                | Refluxing in EtOH, 0.1 g | 6        | 62        |
| Fe3O4@SiO2-pr             | Refluxing in EtOH, 0.1 g | 6        | 64        |
| Fe3O4@SiO2-Pr@dapsone     | Refluxing in EtOH, 0.1 g | 5        | 73        |
| Fe3O4@SiO2-propyl@dapsone-Cu | Refluxing in EtOH, 0.1 g | 3       | 97        |
| Fe3O4@SiO2-propyl@dapsone-Cu | Refluxing in EtOH, 0.01 g | 6       | 76        |
| Fe3O4@SiO2-Pr@dapsone-Cu  | Refluxing in EtOH, 0.05 g | 3       | 96        |
| Fe3O4@SiO2-propyl@dapsone-Cu | Stirring at room temperature in water, 0.05 g | 3     | 98        |
### TABLE 2 | The reaction scope of synthesis of pyranochromene-diones.

| Entry | Product | Time (h) | Yield (%) $^{a,b}$ | Mp (°C) |
|-------|---------|----------|---------------------|---------|
| 1     | ![Product 1](image1.png) | 3        | 98                  | 203–205 |
| 2     | ![Product 2](image2.png) | 3        | 96                  | 176–177 |
| 3     | ![Product 3](image3.png) | 3.5      | 97                  | 276–278 |
| 4     | ![Product 4](image4.png) | 4        | 94                  | 289–290 |
| 5     | ![Product 5](image5.png) | 4        | 95                  | 269–270 |
| 6     | ![Product 6](image6.png) | 4        | 96                  | >300    |
| 7     | ![Product 7](image7.png) | 5        | 94                  | 203–205 |
| 8     | ![Product 8](image8.png) | 5.5      | 90                  | 234–235 |

$^a$Isolated yield.

$^b$All of the synthesized compounds are new and are characterized by FT-IR, NMR, and elemental analysis.
features of the catalyst were not changed after several uses (Figure 6).

To better understand the stability of catalyst after five cycles under these reaction conditions, FE-SEM and TEM analyses were carried out. The results are summarized in Figure 7.

**CONCLUSION**

In conclusion, a new catalytic method for the synthesis of pyranochromene-diones has been developed. This method offers several advantages, such as simple workup and purification procedure without the use of any chromatographic method,
mild reaction conditions, use of inexpensive and commercially available starting materials, recyclability and reusability of the catalyst, high product yields, and short reaction time. So we think that this procedure could be considered a new and useful addition to the present methodologies in this area.

DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article-supplementary files, further inquiries can be directed to the corresponding author.

REFERENCES

Abbasi, Z., Rezayati, S., Bagheri, M., and Hajinasiri, R. (2017). Preparation of a Novel, Efficient, and Recyclable Magnetic Catalyst, γ-Fe2O3@HAp-Ag Nanoparticles, and a Solvent Halogen-free Protocol for the Synthesis of Coumarin Derivatives. Chin. Chem. Lett. 28, 75–82. doi:10.1016/j.ccl.2016.06.022

Barrard, D. L., Xu, Z. Q., Stowell, V. D., Yuan, H., Smee, D. F., Samy, R., et al. (2002). Coumarins and Pyronarocoumarins, Potential Novel Pharmacophores for Inhibition Ofmeasles Virus Replication. Antivir. Chem. Chemother. 17, 39–59. doi:10.1177/0956320202013010104

Bienaymé, H., Hulme, C., Oddon, G., and Schmitt, P. (2000). Maximizing Synthetic Efficiency: Multi-Component Transformations lead the Way. Chem. Eur. J. 6, 3321–3329. doi:10.1002/1521-3765(20000915)6:18<3321::aid-chem3321>3.0.co;2-a

Dömling, A. (2002). Recent Advances in Isocyanide-Based Multicomponent Chemistry. Curr. Opin. Chem. Biol. 6, 306–313. doi:10.1016/S1367-5931(02)00328-9

Dömling, A., and Ugi, I. (2000). Multicomponent Reactions with Isocyanides. Angew. Chem. Int. Ed. 39, 3168–3210. doi:10.1002/1521-3773(20000915)39:18<3168::AID-AJIC3168>3.0.CO;2-a

Fekri, L. Z., and Fard, H. S. (2016). 1,4-Diazaniumbicyclo[2.2.2]octane Diacetate: As an Effective, New and Reusable Media for the Synthesis of 14-Aryl-14h-Benzo[d]imidazo[2,1-a]xanthenes. Acta Chim. Slov. 63, 263–270. doi:10.17344/actsci.2015.2096

Fekri, L. Z., and Maleki, R. (2017). KIT-6 Mesoporous Silica-Coated Magnetic Nanoparticles: A Highly Efficient and Easily Reusable Catalyst for the Synthesis of Benzo[d]imidazole Derivatives. J. Hetero. Chem. 54, 1167–1171. doi:10.1002/jhet.2686

Fekri, L. Z., Nikpassand, M., Pourmousajani, S., and Aghazadeh, B. (2018a). Synthesis and Characterization of Amino pH-sensitive Functionalized Silica-Coated NiFe2O4 Nanoparticles: a Heterogeneous, New and Magnetically Separable Catalyst, high product yields, and short reaction time. So we think that this procedure could be considered a new and useful addition to the present methodologies in this area.

AUTHOR CONTRIBUTIONS

LZ carried out experimental studies, wrote the original draft, and analyzed spectral characterization of synthesized molecules and project planning, proofreading, and editing.

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Ammonium Acetate. Tetrahedron Lett. 53, 3448–3451. doi:10.1016/j.tetlet.2012.04.088

Inaloo, I. D., Esmailpour, M., and Esmaeilpour, M. A. (2020a). Nickel-Catalyzed Synthesis of N-Heteroaryl Carbazoles from Cyanate Salts and Phenols Activated with Cyanuric Chloride. Chem. Cat. Chem. 12 (21), 5486–5491. doi:10.1016/j.cctc.202000876

Inaloo, I. D., Majnooni, S., Eslahi, H., and Esmailpour, M. M. (2020b). Air-Stable Fe3O4@SiO2–EDTA-Ni(0) as an Efficient Recyclable Magnetic Nanocatalyst for Effective Suzuki-Miyaura and Heck. Cross-coupling via aryl Sulphonates and Carbazoles 34, e5662. doi:10.1002/aoc.5662

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Synthesis of Pyranochromene-diones by Novel Nano Catalyst

Nikpassand, M., Zare, L., and R. Mousavi, M. (2012). Comparative Study for the Catalytic Synthesis of Phenols Activated with Cyanuric Chloride. Chem. Cat. Chem. 11, 1091–1095. doi:10.1016/j.cctc.2012.01.001

Nikpassand, M., Fekri, L. Z., and Nabatzadeh, M. (2017). Fe3O4@SiO2@KIT-6 as an Efficient and Reusable Catalyst for the Synthesis of Novel Derivatives of 3,3'-((Aril-1-Phenyl-1h-Pyrazol-4-yl)methylene)bis(1H-Indole). Comb. Chem. High. Throughput. Screen. 20, 533–538. doi:10.2174/13862073206614021523248

Nikpassand, M., Zare, L., and R. Mousavi, M. (2012). Comparative Study for the Aqueous Synthesis of New Generation of Diiodomethanes Using L-Proline, K10 and Nano-Fe3O4 under Ultrasound Irradiation. Loc 9 (5), 375–381. doi:10.2174/157017812801264719

Ou, C. N. H., Selim, Y. A., Sakran, G., and Badr El-din, A. S. (2014). Synthesis of Pyranochromene and Pyranopyrimidine Derivatives from Substituted Natural Coumarin Isolated from Ammi Majus L. And Their Biological Evaluation. Med. Chem. Res. 23, 1180–1188. doi:10.1007/s00044-013-0724-z

Rezaei Nezhad, E., Sajjadifar, S., Abbasi, Z., and Rezayati, S. (2014). Chemoselective Synthesis of 1.1-diacetate Using Ni(II)hydroxyapatite-core@shell γ-Fe2O3 Nanoparticles as an Efficient and Reusable Lewis Acid Catalyst under Solvent Free Conditions. J. Sci. 25, 125–132.

Rezayati, S., Jafroudi, M. T., Nezhad, E. R., Hajmarsi, R., and Abbaspour, S. (2016). Imidazole-functionalized Magnetic Fe3O4 Nanoparticles: An Efficient, Green, Recyclable Catalyst for One-Pot Friedländer Quinoline Synthesis. Res. Chem. Intermed. 42, 5887–5898. doi:10.1007/s11164-015-2411-9

Sardarian, A. R., Dindarloo, Inaloo, L., and Zangiabadi, M. (2019). An Fe3O4@SiO2/Schiff base Cu(ii) Complex as an Efficient Recyclable Magnetic Nanocatalyst for Selective Mono N-Arylation of Primary O-Alkyl Thio-carbamates and Primary O-Alkyl Carbazoles with Aryl Halides and Arylboronic Acids. New J. Chem. 43, 8557–8565. doi:10.1039/C9NJ00028C

Su, C.-R., Liu, C. M., Damu, A. G., Kuo, T.-H., Chiang, P.-C., et al. (2009). Anti-HBV and Cytoxic Activities of Pyranocoumarin Derivatives. Bioorg. Med. Chem. 17, 6137–6143. doi:10.1016/j.bmc.2008.12.007

Symeonidis, T., Fylaktakidou, K. C., Hadjipavlou-Litina, D. J., and Litinas, K. E. (2009). Synthesis and Anti-inflammatory Evaluation of Novel Angularly or Linearly Fused Coumarins. Eur. J. Med. Chem. 44, 5012–5017. doi:10.1016/j.ejmech.2009.09.004

Williams, R. M., and Cox, R. J. (2003). Paraherquamides, Brevaniamides, and Asperparalines: Laboratory Synthesis and Biosynthesis. An Interim Report. Acc. Chem. Res. 36, 127–139. doi:10.1021/ar020229e

Xu, Z.-Q., Pupek, K., Suling, W. J., Enache, L., and Flavin, M. T. (2006). Pyranocoumarin, a Novel Anti-TB Pharmacophore: Synthesis and Biological Evaluation against Mycobacterium tuberculosis. Bioorg. Med. Chem. 14, 4610–4626. doi:10.1016/j.bmc.2006.02.017

Zare, L., Mahmoodi, N. O., Yahyazadeh, A., Mamaghani, M., and Tabatabaeean, K. (2010). An Efficient Chemo- and Regioselective Three-Component Synthesis of Pyridazine and Phthalazinones Using Activated KSF. Chin. Chem. Lett. 21, 538–541. doi:10.1016/j.jcl.2009.11.032

Zhang, G., Zhang, Y., Yan, J., Chen, R., Wang, S., Ma, Y., et al. (2012). One-pot Enantioselective Synthesis of Functionalized Pyranocoumarins and 2-Amino-4h-Chromenes: Discovery of a Type of Potent Antibacterial Agent. J. Org. Chem. 77, 878–888. doi:10.1021/jo202020m