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FULL PAPER

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Preparation of magnetically recyclable ZnFe₂O₄ nanoparticles by easy single-step co-precipitation method and their catalytic performance in the synthesis of 2-aminothiophenes

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R. Tayebee, Department of Chemistry, Hakim Sabzevari University, Sabzevar, 96179-76487, Iran. Email: rtayebee@hsu.ac.ir In this work, a new synthetic route for the preparation of $ZnFe_2O_4$ nanoparticles through the chemical co-precipitation using Fe^{2+} and Fe^{3+} ions in an alkaline solution was developed. The synthesized nanoparticles were characterized by XRD, FTIR, SEM, ICP-MS, DRS, TGA, VSM and elemental analysis. Characterization results confirmed the formation of single $ZnFe_2O_4$ phase, with an average particle size of 40 nm and a high saturation magnetization of 34 emu g^{-1} . The prepared material was employed as a catalyst for the synthesis of 2-aminotiophene derivatives through the Gewald reaction. This thermally and chemically stable nanocatalyst is environmentally benign, economical and reusable which can be easily recovered using an external magnet. Therefore, it appears that this methodology can be simply extended for industrial purposes.

KEYWORDS

2-aminothiophenes, Gewald reaction, magnetic nanoparticles, single-step co-precipitation, ZnFe₂O₄

1 | INTRODUCTION

Synthesis and application of ferrite nanoparticles of the type MFe₂O₄, where M stands for divalent metal ions such as Zn, Co, Mn, Mg, Ni, etc. have attracted extensive consideration in the last decades.^[1,2] Ferrites are ceramic materials possessing interesting magnetic properties, excellent chemical stability and mechanical strengths, and offering widespread applications in several industrial segments.^[2] Among them, ZnFe₂O₄ has received much attention due to its potential uses in optical and electrical devices,^[3] as adsorbent material for hot-gas desulfurization,^[4] in catalytic transformations,^[5] and in biomedicine.^[6] ZnFe₂O₄ has been fabricated using different procedures, such as co-precipitation,^[7] solid-state reaction,^[8] sol-gel,^[9] glycine combustion method,^[10] microwave-assisted solvothermal and solvothermal synthesis,^[11] combustion reaction using urea as reducing

agent,^[12] high energy ball-milling,^[13] phase chemical reaction,^[14] polyethylene glycol-assisted route,^[15] thermal plasma synthesis,^[16] one-step solid microwave combustion method,^[17] and synthesis in supercritical fluids.^[18] Zinc ferrite can be separated from reaction mixtures by applying an external magnet and could be reused in further reactions.^[19,20]

The improvement of new methodologies for the direct synthesis of significant complex molecules is a focal task for current organic chemists.^[21] In this unique situation, improvement of multi-component reactions is of fundamental significance.^[22] 2-Aminothiophenes have been increasingly used as antitumor and antibacterial agents, antioxidants, dyes, pharmaceutical and agrochemical compounds.^[23] Additionally, the thiophene ring is likewise found in numerous natural and synthetic biologically active compounds.^[24] In this context, 2-aminothiophene synthesis has attracted considerable attention, and many

2 of 7 WILEY-Organometallic Chemistry

synthetic protocols have been developed.^[24] However, many of the reported methodologies require troublesome preparation of the starting materials, multistep synthesis, and resulting yields are low.

As a result of our current research and in continuation of our previous works,^[25] we are going to present an eco-friendly, novel, and low-cost route for the synthesis of $ZnFe_2O_4$ nanoparticles using a single-step coprecipitation method under green and environmentally friendly conditions. The obtained $ZnFe_2O_4$ nanoparticles are then utilized as an efficient and reusable catalyst in the synthesis of different 2-aminothiophenes based on the Gewald reaction in solvent-less conditions at 100 °C.^[26]

2 | EXPERIMENTAL

2.1 | Materials

All chemicals were purchased from Sigma-Aldrich and used without further purification: Ferrous chloride tetrahydrate (FeCl₂.4H₂O), ferric chloride hexahydrate (FeCl₃.6H₂O), zinc oxide (ZnO) and ammonium hydroxide (NH₄OH; 28%).

2.2 | Preparation of ZnFe₂O₄

Zinc ferrite nanoparticles were obtained by the dispersion of 0.2 g of ZnO nanoparticles in a 20 ml solution of deionized water including FeCl₃.6H₂O (0.54 g; 2 mmol) and FeCl₂.4H₂O (0.198 g; 1 mmol) at 50 °C under magnetic agitation and nitrogen atmosphere. After that, the mixture was stirred for 10 min; then, an aqueous solution of NH₄OH (10 ml, 8 M) was slowly added to start the precipitation of ZnFe₂O₄. In order to accelerate the growth of the particles, the reaction was performed at 50 °C for 10 min by magnetic stirring. Finally, the precipitate was separated by a magnetic field and washed thoroughly with deionized water for three times. Subsequently, the produced solid was dried at 80 °C for 12 h. To study impact the of drying conditions on the catalytic efficacy of ZnFe₂O₄, we dried the prepared final material under freeze conditions. Conclusions demonstrated no obvious enhancement in the catalytic activity of this sample compared with the one dried under conventional thermal conditions as described above.

2.3 | Characterization techniques

Field emission scanning electron microscopy (FESEM) was performed with XL-30 Phillips (1992). The purity of the sample was assessed by X-ray diffraction (XRD) analysis on a PW1800-PHILIPS diffractometer with Cu-K_{α}

radiation ($\lambda = 1.5418$ Å) at 40 keV and 40 mA. The Fourier transform infrared (FTIR) spectrum was recorded on an 8700 Shimadzu spectrophotometer utilizing KBr pellets. ¹H- and ¹³C-NMR spectra were recorded on a Bruker AVANCE 300 MHz spectrometer. Diffuse reflection/transmittance spectroscopy (DRS/DTS) was acquired on an Avaspec-2048-TEC equipment. The chemical composition of the prepared material was obtained using an inductively coupling plasma spectrometer (ICP-MS; model Vista-pro). Thermal gravimetric analysis (TGA) was done on a Bahr STA-503 instrument on air atmosphere at a heating rate of 10 °C min⁻¹. Magnetic property of the catalyst was attained via vibrating sample magnetometer/alternating gradient force magnetometer (VSM/ AGFM, MDK Co, Kashan, Iran, www.mdk magnetic. com). A freeze dryer (Model FD-10, Pishtaz Equipment Engineering Co, Tehran, Iran) was utilized for drying of the sample. Elemental analyses were accomplished with a Thermo Finnigan Flash-1112EA microanalyzer. The progress of the reactions was shown by thin-layer chromatography (TLC). All the synthesized 2-aminothiophenes were distinguished by comparison of their physical and spectral information with those already announced.^[27-30]

2.4 | Synthesis of 2-aminothiophene derivatives

5 mmol of malonodinitrile, 5 mmol of sulfur powder and 5 mmol of ketone, were thoroughly mixed. Then, 0.03 g of $ZnFe_2O_4$ nanoparticles was added, and the mixture was warmed to 100 °C with constant stirring for 4 h. Eventually the reaction was stopped, $ZnFe_2O_4$ was separated with a simple magnet, and the final product was dissolved in 10 ml of hot ethanol. After that, the reaction mixture was cooled to room temperature and decanted into 100 ml of ice-water. The precipitate was filtered, washed with cold water (5 °C) and dried at room temperature for 12 h. To continue refinement, we purified the crude product via silica gel column chromatography with 10: 1 hexane: ethyl acetate as eluent. Scheme 1 displays a reaction scheme employed in this work for the synthesis of 2-aminothiophenes.



SCHEME 1 Reaction scheme for the synthesis of 2-aminothiophenes

2.5 | Spectral data of some selected compounds^[30]

2.5.1 | 2-Amino-5-ethylthiophene-3carbonitrile

FT-IR (KBr) (ν_{max}/cm^{-1}): 3440, 3360, 3220, 2960, 2220, 1650, 1560, 1460, 1380, 1080, 840. ¹H–NMR (300 MHz, CDCl₃): δ_{H} (ppm) 6.34 (s, 1H), 4.73 (brs, 2H), 2.60 (q, J.7.2 Hz, 2H), 1.20 (t, J.7.2 Hz, 3H); ¹³C–NMR(CDCl₃): δ 161.0, 132.0, 120.2, 115.9, 86.7, 23.0, 15.2 ppm.

2.5.2 | 2-Amino-4,5,6,7-tetrahydro-1benzothiophene-3-carbonitrile

FT-IR (KBr) (ν_{max} /cm⁻¹): 3440, 3340, 3200, 2910, 2840, 2200, 1650, 1640, 1520,1320, 1120, 680; ¹H–NMR (300 MHz, CDCl₃): δ_{H} (ppm) 1.78 (t, 4H,2CH₂), 4.64 (s, 2H, NH₂), 2.49 (m, 4H, 2CH₂); ¹³C–NMR(CDCl₃): δ 22.10, 23.34, 23.72, 24.10, 88.45, 115.59, 120.50, 132.27,160.15 ppm.

2.5.3 | 2-Amino-5, 6, 7, 8-tetrahydro-4Hcyclohepta[b]thiophene-3-carbonitrile

FT-IR (KBr) (ν_{max}/cm^{-1}): 3440, 3309, 3207, 2921, 2842, 2200, 1622, 1569, 1514, 1438, 1400, 1344, 1128, 829, 493. ¹H–NMR (300 MHz, CDCl₃): δ_{H} (ppm) 4.58 (s, 2H), 2.56–2.63 (m, 4H), 1.80–1.82 (m, 2H), 1.64–1.65 ppm (m, 4H); ¹³C–NMR(CDCl₃): δ 158.2, 136.8, 123.7, 116.0, 91.7, 31.9, 29.4, 29.1, 28.1, 27.2 ppm.

2.5.4 | 2-Amino-4-phenylthiophene-3carbonitrile

FT-IR (KBr) (ν_{max}/cm^{-1}): 3420, 3320, 3200, 2220, 1630, 1570, 1430, 1300, 1190,940, 768, 715; ¹H–NMR (300 MHz, CDCl₃): δ_{H} (ppm) 7.60 (m, 2H), 7.44 (m, 2H), 7.38 (m, 1H), 6.32 (s, 1H), 5.02 (brs, 2H); ¹³C–NMR(CDCl₃): δ 164.1, 139.9, 134.2, 128.8, 128.2, 127.2, 116.2, 105.9, 88.0 ppm.

3 | **RESULTS AND DISCUSSION**

3.1 | Characterization

XRD was used to investigate the phase of produced binary, $ZnFe_2O_4$, oxide. As shown in Figure 1, the XRD pattern displayed diffraction lines of a high crystalline nature at $2\theta = 30.0$, 35.2, 42.9, 53.2 and 56.5° , which corresponds to (220), (311), (400), (422), and (511) crystal planes, respectively. This pattern matched well with the JCPDS of 22–1012 confirming the



FIGURE 1 XRD pattern of ZnFe₂O₄

formation of ZnFe₂O₄ structure.^[31] Moreover, no impurity peaks were observed, confirming the high purity of the sample.

The FTIR spectrum of $ZnFe_2O_4$ is shown in Figure 2. Due to the interactions between oxygens and cations in the octahedral and tetrahedral environment; three vibrational frequencies were shown in the spinel structure of zinc ferrite (666 cm⁻¹, 543 cm⁻¹, 450 cm⁻¹). The major frequency bands 666 and 543 cm⁻¹ correspond to vibrations of the metal at the tetrahedral site (tetrahedral Zn^{2+} stretching), and the low-frequency band 453 cm⁻¹ is proven with Fe³⁺ vibration frequencies in the octahedral sites.^[32] The weak band close to 1590 cm⁻¹ is assigned to H–O–H bending vibration mode due to the absorption of humidity, because the FT-IR sample disks were prepared in an open-air atmosphere. The band at 3400 cm⁻¹ corresponds to the attendance of hydroxyl groups.^[33]



FIGURE 2 FTIR spectrum of ZnFe₂O₄ nanoparticles

4 of 7 WILEY-Organometall Chemistry

Optical properties of $ZnFe_2O_4$ were investigated by UV–Vis diffuse spread reflectance (DRS) measurement at room temperature (Figure 3). It is explicit that the obtained sample shows excellent visible-light absorption, which is consistent with the black color of $ZnFe_2O_4$. The high visible light absorbance demonstrates that the obtained $ZnFe_2O_4$ may have high visible-light utilization efficiency. Besides, the absorption band of $ZnFe_2O_4$ shows a relatively steep edge without a shoulder, illustrating that the absorption of the visible light range can be assigned to a characteristic band transition instead of surface states.^[34]

Distribution and morphology of $ZnFe_2O_4$ particles was investigated by FESEM (Figure 4). Qualitative analysis *via* FESEM revealed a homogeneous and uniform distribution of spherical $ZnFe_2O_4$ particles, with an average size of 40 nm. Chemical composition (mol%) of the $ZnFe_2O_4$ sample was obtained from ICP analysis, being Zn and Fe of 14.20 and 28.49, respectively, resulting in the atom ratio of 1:2. This finding is in fair agreement with the atomic composition of $ZnFe_2O_4$, thus confirming the formation of single-phase $ZnFe_2O_4$.

The magnetic property of $ZnFe_2O_4$ was characterized by VSM (Figure 5). Detailed magnetic measurements, i.e., zero-field cooling and magnetic hysteresis loops at several temperatures, field cooling magnetization vs temperature, relaxation under different applied fields at 5 K, have been accomplished to check the magnetic property of the synthesized $ZnFe_2O_4$ nanoparticles. The superparamagnetic property of the catalyst, which accounts for its simple recovery, was corroborated by VSM analysis. There was no hysteresis loop, and the ratio of magnetic remanence/ magnetic saturation was 34.0 emu g⁻¹.^[35,36] The saturation magnetization of spinel ferrites originates from the difference in the magnetic moments of the cations occupying the octahedral and tetrahedral lattice sites positions. As Zn^{2+} cation is not magnetic, its value for Zn-based ferrites



FIGURE 3 DRS spectrum of ZnFe₂O₄



FIGURE 4 SEM images of ZnFe₂O₄



FIGURE 5 Magnetization vs applied magnetic field for ZnFe₂O₄ nanoparticles

directly reflects the distribution of the Fe³⁺ ions among two sub-lattices.^[37,38]

The thermal stability of the prepared $ZnFe_2O_4$ nanoparticles in air was investigated (Figure 6). According to the TGA analysis, a distinct weight gain (~2.6%) was



FIGURE 6 Thermogravimetric (TGA) analysis for ZnFe₂O₄

occurred due to absorption of oxygen atom. This finding clearly confirmed zinc ferrite was started to decompose at 400 °C with a distinct mass loss (around 4%). Furthermore, continues mass loss around 90–200 °C is attributed to the loss of surface moisture. Notably, release of surface moisture was masked with the mentioned weight gain. To study the thermal stability of the prepared $ZnFe_2O_4$, it was calcined in a muffle-oven at 600 °C for 3 h. Color change of zinc ferrite to brown was obvious. XRD analysis of the final material showed that ZnO and Fe_3O_4 were generated as minor phases; however, $ZnFe_2O_4$ was observed as the major phase. This finding clearly confirmed the TGA analysis.

3.2 | Catalytic activity of ZnFe₂O₄ in the synthesis of 2-aminothiopohene derivatives

The catalytic effectiveness of the heterogeneous catalytic system was checked for the synthesis of 2-amino-4,5,6,7-tetrahydro-1-benzothiophene-3-carbonitrile with different

TABLE 1 Effect of ZnFe₂O₄ amount on the condensation of cyclohexanone, elemental sulfur and malonodinitrile.

Entry	ZnFe ₂ O ₄ (g)	ZnFe ₂ O ₄ (mol%)	Time (h)	Yield (%)
1	0	0	4	6
2	0.01	0.85	4	57
3	0.02	1.65	4	60
4	0.03 ^a	2.50	4	80
5	0.04	3.30	4	60
6	0.05	4.10	4	52

Reaction conditions: malonodinitrile (5 mmol), Cyclohexanone (5 mmol), and sulfur powder (5 mmol) were mixed in the presence of nano-ZnFe₂O₄ and the reaction mixture was warmed to 100 °C for 4 h.

^aNo obvious enhancement on the yield was observed when the catalyst treated under freeze drying was used.



FIGURE 7 Effect of reaction time on the condensation reaction

amounts of ZnFe₂O₄. Thus, the reaction of elemental sulfur, cyclohexanone, malonodinitrile was investigated under a solventless condition at 100 °C in the presence of 0–0.03 g of nanocatalyst. The screening results of the reaction are summarized in Table 1. In the absence of the catalyst, the reaction was rather non-profitable, and just 6% of the product was attained after 4 h (entry 1). On the other hand, the addition of 0.01 g (0.85 mol%) and 0.03 g (2.5 mol%) of the catalyst provided 57 and 80% yield, respectively, during the same time (Entries 2 and 4). As expected, the yield was growing with increasing the catalyst concentration. However, excessive amounts of the catalyst lowered the yield to 52% (Entry 6). Such lowering can be explained by the decreasing concentration of reactants

TABLE 2Synthesis of different 2-aminothiophene derivativesunder the optimum reaction conditions.

Entry	Ketone/aldehyde	Product	M.P.	Yield(%)
1	∧ ↓	NH ₂	120-122	30 ^a
2			144–145	55
3			146–147	80
4		CN SNH2	128–130	51
5		NC NH2	134–136	35
6			148-150	25

^aIn absolute ethanol (4 h reflux). General reaction condition is described in the footnote of Table 1.

TABLE 3 Comparison of the catalytic activity of $ZnFe_2O_4$ with some reported catalysts in the synthesis of 2-amino-4,5,6,7-tetrahydro-1-benzothiophene-3-carbonitrile.

Entry	Catalyst	Time (h)	Tomp (°C)	Viold (97)	Condition/	Cat.	E Eastor (g)	TON	Dof
Linuy	Catalyst	Time (II)	Temp. (C)	11eiu (<i>%</i>)	Solvellt	(g)	E-Factor (g)	IUN	Kel.
1	$ZnFe_2O_4$	4	100	80	free	0.03	0.24	2667	This work
2	ZnO _{34%} /NCP	4	100	76	free	0.05	6.30	1520	[39]
3	Bulk TiO ₂	10	100	74	free	0.04	6.52	1850	[27]
4	Bulk ZnO	10	100	65	free	0.04	7.60	1625	[27]
5	Nano ZnO	6	100	70	free	0.02	6.95	3500	[39]
6	MgO/Al ₂ O ₃	8	60	91	ethanol	0.05	17.04	1820	[40]
7	KF/Al ₂ O ₃	3.5	78	91	ethanol	0.2	15.84	455	[41]

Turnover number (TON) was calculated as yield(%)/g of catalyst.

at the active sites. Hence, 0.03 g of catalyst was determined as the optimal quantity for the reaction.

Figure 7 demonstrates the effect of reaction time on the yield under the standard reaction condition. It was observed that the yield grows with augmenting the reaction time.

To focus the universality of this catalyst, we examined cyclopentanone, cyclohexanone and cycloheptanone, as cyclic ketones, along with butyraldehyde, diethyl ketone and acetophenone. All the examined carbonyl compounds showed good reactivity towards the Gewald condensation reaction and led successfully to the desired products with good selectivity (Table 2). However, findings revealed that linear carbonyl compounds were less efficient than cyclic ketones. Moreover, cyclohexanone provided the best yield and behaved better than cyclopentanone and cycloheptanone.

Several protocols have been accounted for the synthesis of 2-amino-thiophenes. The reaction efficiency of the present method was compared with those reported in the literature, and the results are summarized in Table 3. The comparison was performed regarding catalyst amount, reaction time and yield. The obtained results demonstrated that 0.03 g of ZnO prompted to 33% of the desired product; while, Fe₃O₄ was ineffectual after 4 h. Meanwhile, ZnFe₂O₄ showed a high activity and led to 80% yield. The oxide anions, as the basic parts of the catalyst, would catalyze addition and cyclization of the condensation reactions; whereas, the dehydration may proceed by Fe^{3+} of Fe_2O_3 as the acidic parts of the catalyst.^[31] Except the commercial ZnO, all the solid bases can produce the desired product in convenient yields under moderate conditions. However, our methodology offers a one pot single-step preparation of the target products under aerobic, simple, and solvent-free conditions without utilizing the powerful Lewis bases. Moreover, the present process allows some advantages such as mild reaction condition, environmentally benign nature of the catalyst, generality, small amount of the additive, and economical production.

4 | CONCLUSION

In this study, ZnFe₂O₄ nanoparticles have been successfully prepared using a rapid and facile method through the chemical co-precipitation route of Fe²⁺ and Fe³⁺ ions in an alkaline medium. Then, the prepared ZnFe₂O₄ magnetic nanoparticles were used as an environmentally friendly heterogeneous catalyst for the synthesis of 2aminothiophene derivatives by Gewald reaction. We showed that the high activity of zinc ferrite could originate from the dual Lewis acidic Fe³⁺ sites along with oxide and Zn^{2+} ions, which can act as Lewis basic centers. The present methodology is characterized by sufficient generality, compatibility with different functional groups, short reaction time, high yield, easy work-up, and environmentally benign conditions. Owing to the reusing and recycling of the catalyst, the method might solve many separation problems it could be easily scaled-up in organic synthesis as well as in industry.

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Applied

Organometallic

7 of 7

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