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FULL PAPER

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Design and preparation of ZnS-$ZnFe_2O_4$: a green and efficient hybrid nanocatalyst for the multicomponent synthesis of 2,4,5-triaryl-1*H*-imidazoles

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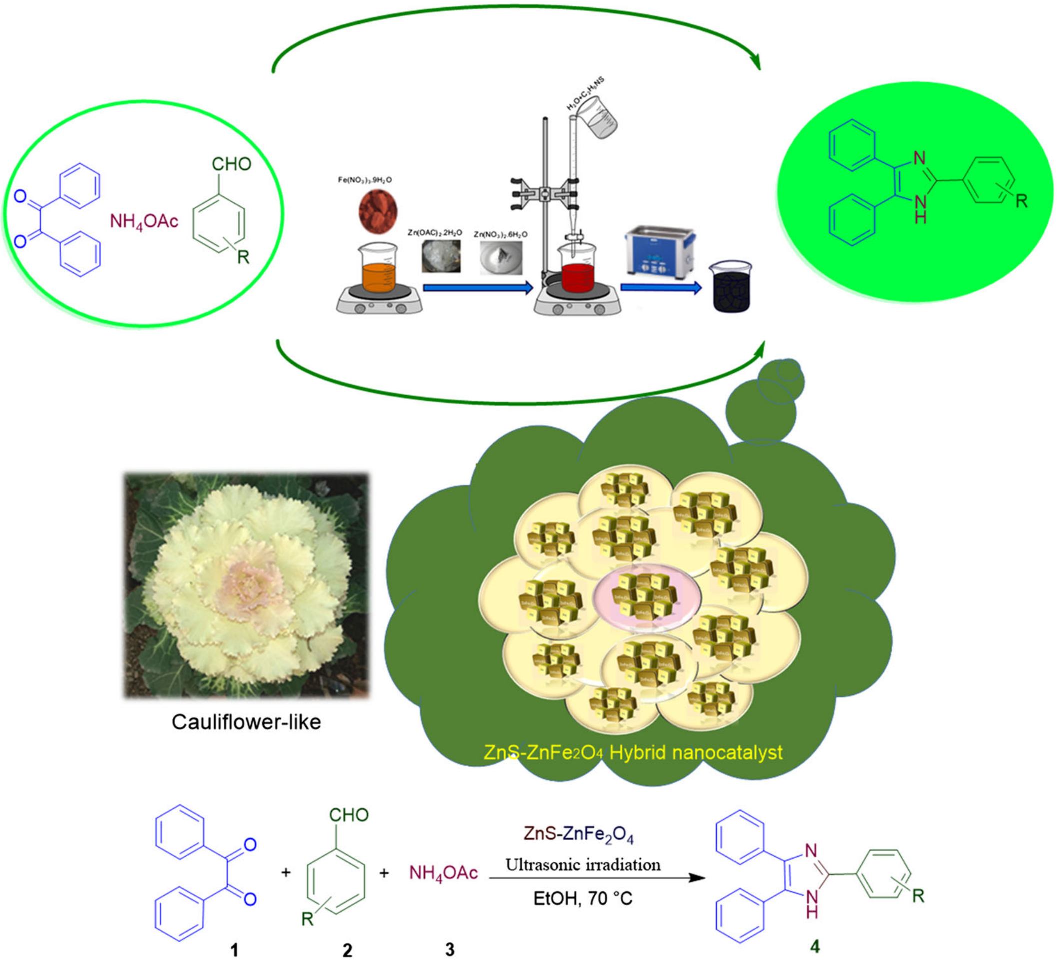
In the present work, a new protocol was introduced for the preparation of an efficient hybrid nanocatalyst ZnS-$ZnFe_2O_4$ via the co-precipitation method as well as its application in the synthesis of 2,4,5-triaryl-1*H*-imidazoles derivatives starting from various aromatic aldehydes, benzil and ammonium acetate under ultrasonic irradiation in ethanol. ZnS-$ZnFe_2O_4$ was characterized by Fourier transform infrared (FT-IR) spectroscopy, energy-dispersive X-ray spectroscopy (EDS) analysis, scanning electron microscopy (SEM) image, X-ray diffraction (XRD) pattern and vibrating sample magnetometer (VSM) curve. This method has advantages such as high efficiency of the heterogeneous catalyst, the use of environmentally-friendly solvent, high yields, short reaction times and easy isolation of the products and chromatography-free purification. Our outcomes illustrated that the present nanocatalyst with nearly spherical and Cauliflower-like morphology and average particle size of 36 nm could be applied as an effective and magnetically recyclable catalyst without any significant decreasing of activity. Furthermore, the synergic effect of bimetallic Lewis acids was studied for the synthesis of imidazole derivatives.

KEYWORDS
green chemistry, heterogeneous nanocatalyst, imidazole, recyclable, three-component reaction

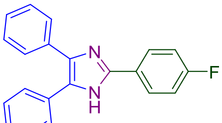
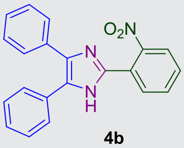
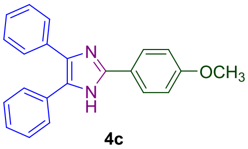
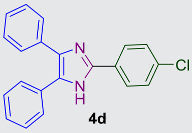
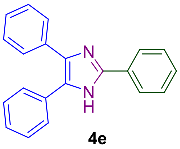
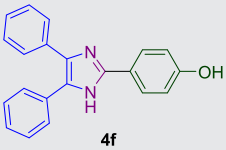
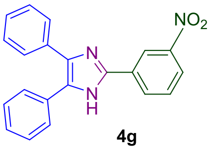
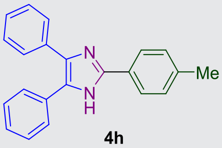
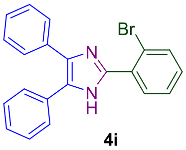
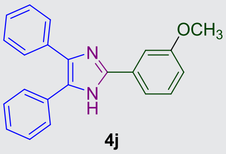
1 | INTRODUCTION

In recent years, mixed transition metal oxides (MTMOs), compounded by two simple TMOs, have attracted much attentions both in academic and industrial researches. Categories of MTMOs include MMn_2O_4 (M = Zn, Ni and Co), MCo_2O_4 (M = Mn, Ni, Zn and Fe) and MFe_2O_4 (M = Co, Ni, Mn and Zn).[1] Due to considerable magnetic properties of spinel-type complexes of MFe_2O_4 has widely been used in various fields such as magnetic resonance imaging (MRI), biomolecular sensors,[2] electronic storage, frequency core, data storage and drug delivery.[3–5] Spinel nanocrystalline zinc ferrites have been applied in various organic reactions as effective photocatalysts.[6] $ZnFe_2O_4$ is an important MTMO semiconductor with a narrow band gap.[7] In cubic ferrites like MFe_2O_4, in which M is a metal cation, the structural oxygen is located in an FCC compact network and the M^{+2} and Fe^{+3} are located in empty hollow tetrahedral and octahedral. Zinc ferrite has widely been used in various fields including magnetic fluids for the storage information,[8] photocatalysis[9] and MRI.[10] ZnS nanoparticles are excellent semiconductors with an energy level of 3.8 ev and have two crystalline structures: hexagonal wurtzite and cubic zinc blend. In the cubic structure of ZnS, there are three axes of x, y and z in cubic crystal structure.[11] ZnS nanoparticles have been analyzed for their superior performance as a catalyst. The enhancement of efficient techniques for the synthesis of complex molecules is one of the essential assignment for organic chemists. In this regard, the multicomponent reactions (MCRs) are potentially important approaches. Although, the synthesis of imidazole core as a major component of many drugs is of prime importance. In addition, they have various medicinal and biological activities like antibacterial,[12] antifungal,[13] reducing inflammation,[14] anti-cancer[15] antiviral,[16] anti-diabetic,[17] anti-allergic,[18] analgesic[19] and herbicidal.[20] Therefore, in this work, 2,4,5-triaryl-1*H*-imidazole derivatives were synthesized via one-pot and inexpensive method in the presence of ZnS-$ZnFe_2O_4$ MTMO.[21] Disadvantages of previous methods of $ZnFe_2O_4$ preparation include the need for complex equipment, high temperature and energy consumption, low efficiency, isolation problems, as well as long reaction times to complete crystallization of ferrite nanostructures.[22] On the other hand, many of the methods for the synthesis of imidazoles had restrictions like low yields, long reaction times, expensive catalysts, harsh isolation and purification. Synthesis under mild conditions using a suitable, non-toxic, clean and environmentally-friendly and recoverable reagents and catalysts is one of the important goals of green chemistry. As a result, finding an efficient way to prepare these compounds is important.[23] In this work, ZnS-$ZnFe_2O_4$ MTMO as a magnetic recyclable nanocatalyst was prepared and characterized. The results showed that ZnS-$ZnFe_2O_4$ MTMO has not only better catalytic performance in comparison with both ZnS and $ZnFe_2O_4$, but also, could be recovered simply by an external magnet and reused several times in organic reactions. Therefore, the co-precipitation method was applied to control the morphology of the ZnS-$ZnFe_2O_4$ MTMO. The present investigation has various superiority in the preparation of ZnS-$ZnFe_2O_4$ MTMO nanocatalyst to exhibit enhanced nanocatalytic activity and easy magnetic separation via the co-precipitation method[24] as well as environmentally-friendly conditions for the synthesis of diverse 2,4,5-triaryl-1*H*-imidazoles via MCRs under ultrasonic irradiation in ethanol at 70 °C (Scheme 1).



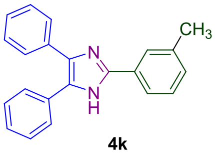
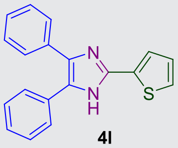
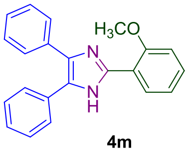
SCHEME 1 Preparation of ZnS-$ZnFe_2O_4$ and synthesis of 2,4,5-triaryl-1*H*-imidazoles in the presence of ZnS-$ZnFe_2O_4$ MTMO.

TABLE 1 Synthesis of 2,4,5-triaryl-1*H*-imidazoles by ZnS-$ZnFe_2O_4$ MTMO under ultrasonic irradiation conditions[a]

Entry	Product	Time (min)	Yield[b] (%)	M.p. (°C) Found/Reported
1	 **4a**	60	68	197–199/196–198[25]
2	 **4b**	50	62	229–231/230–231[26]
3	 **4c**	25	85	229–231/230–231[20]
4	 **4d**	15	95	258–260/259–262[27]
5	 **4e**	32	70	275–277/273–275[28]
6	 **4f**	44	75	257–259/257–258[29]
7	 **4g**	35	92	264–266/265–266[26]
8	 **4h**	30	86	230–231/230–231[20]
9	 **4i**	20	88	199–201/200–201[29]
10	 **4j**	15	95	259–260/258–260[28]
11		45	51	298–299/296–299[30]

(Continues)

TABLE 1 (Continued)

Entry	Product	Time (min)	Yield[b] (%)	M.p. (°C) Found/Reported
	 4k			
12	 4l	45	60	266/264–265[26]
13	 4m	38	66	210–211/210–212[31]

[a] Reaction conditions: benzil (1 mmol), aldehyde (1 mmol), ammonium acetate (4 mmol), EtOH (1 ml) and ZnS-ZnFe$_2$O$_4$ (20 mol%).

[b] Isolated yields.

2 | EXPERIMENTAL SECTION

2.1 | Materials and instruments

All commercially available solvents and chemicals were provided from Aldrich and Merck. Melting points of the samples were measured by an Electrothermal 9100 apparatus. The FT-IR spectra were recorded using a Shimadzu IR-470 spectrometer by using KBr pellet. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker DRX-500 Avance spectrometer at 500 and 125 MHz. VSM curves were measured by LBKFB. SEM images were obtained by the MIRA3 TESCAN instrument. Elemental analysis was performed by using EDS analysis that were obtained by SAMx. The XRD pattern was taken with the Panalytical X′ Pert Pro diffractometer with Cu Kα radiation (40 kV, 40 mA) from 20° to 80° (2θ). All products were identified by comparing of their spectral and analytical data with valid samples. Ultrasonic irradiation were carried out in Elmasonic S60H.

2.2 | Preparation of the ZnS-ZnFe$_2$O$_4$ MTMO hybrid nanocatalyst

Zn (NO$_3$)$_2$·6H$_2$O (0.3 g, 1 mmol), Zn (OAc)$_2$·2H$_2$O (0.2 g, 1 mmol) and thioacetamide (0.1 g, 1.6 mmol) were added to 10 ml of deionized water solution including Fe (NO$_3$)$_3$·9H$_2$O (0.8 g, 2 mmol) during 30 min. Then, the resulting mixture was sonicated. After that, 10 ml of an aqueous solution of NH$_4$OH was added dropwise into the mixture. Eventually, the retrieved precipitate was extracted by an external magnet and washed thoroughly with deionized water and ethanol for two times. Finally, the precipitate was dried at room temperature for 10 hr to provide ZnS-ZnFe$_2$O$_4$ MTMO.

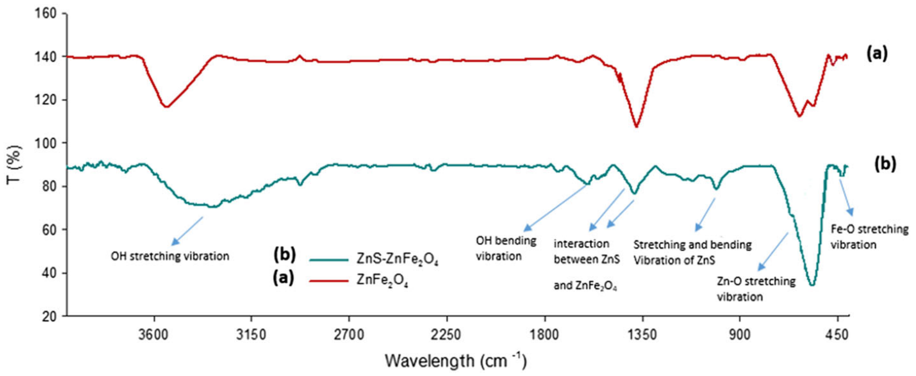


FIGURE 1 FT-IR spectra of: (a) ZnFe$_2$O$_4$ and (b) ZnS-ZnFe$_2$O$_4$ MTMO

2.3 | General procedure for the preparation of 2,4,5-triaryl-1*H*-imidazole derivatives

At first, 4-chlorobenzaldehyde (0.14 g, 1 mmol), ammonium acetate (0.3 g, 4 mmol), benzil (0.21 g, 1 mmol) and ZnS-$ZnFe_2O_4$ nanocatalyst (0.02 g) were stirred at 70 °C in ethanol. The reaction was monitored by thin layer chromatography. After completion of the reaction, the precipitate was washed with deionized water and dried. Pure precipitates were obtained by re-crystallization by EtOH. In all cases, the melting points of the products were measured to be in accordance with reference samples (Table 1).

2.4 | Chemical characterization data of the selected products

2.4.1 | 2-(4-Methylphenyl)-4,5-diphenyl-1*H*-imidazole (4 h)

MP: 230–231 °C; Yield: 86%; FT-IR (KBr, cm^{-1}): 696, 767, 825, 1446, 1496, 1600, 3033, 3380; ^1H NMR ($CDCl_3$): δ_H (ppm); 2.39 (s, 3H, CH_3), 7.24–7.73 (m, 12H, Ar-H), 7.77–7.79 (d, 2H, Ar-H), 9.64 (s, 1H, N-H); ^{13}C NMR ($CDCl_3$): δ_C (ppm); 146.6, 139.2, 130.0, 129.0, 128.2, 127.5, 125.6, 21.8.

2.4.2 | 2-(4-Chlorophenyl)-4,5-diphenyl-1*H*-imidazole (4d)

Mp: 258–260 °C; Yield: 95%; FT-IR (KBr, cm^{-1}): 1448, 1485, 1602, 3060, 3272; ^1H NMR (DMSO-d_6): δ_H (ppm); 7.21–7.56 (m, 12H, Ar-H), 8.11 (d, 2H, Ar-H), 12.79 (s, 1H, N-H); ^{13}C NMR (DMSO-d_6): δ_C (ppm); 144.3, 137.2, 134.9, 132.5, 130.81, 129.1, 128.7, 128.5, 128.4, 128.3, 128.1, 127.0, 126.5, 126.2.

3 | RESULTS AND DISCUSSION

The performance and synthesis of the ZnS-$ZnFe_2O_4$ MTMO were accomplished for the first time by procedures described in this literature. For preparation, in this work, first of all, we presented a simple method for the synthesis of the ZnS-$ZnFe_2O_4$ MTMO as bimetallic Lewis

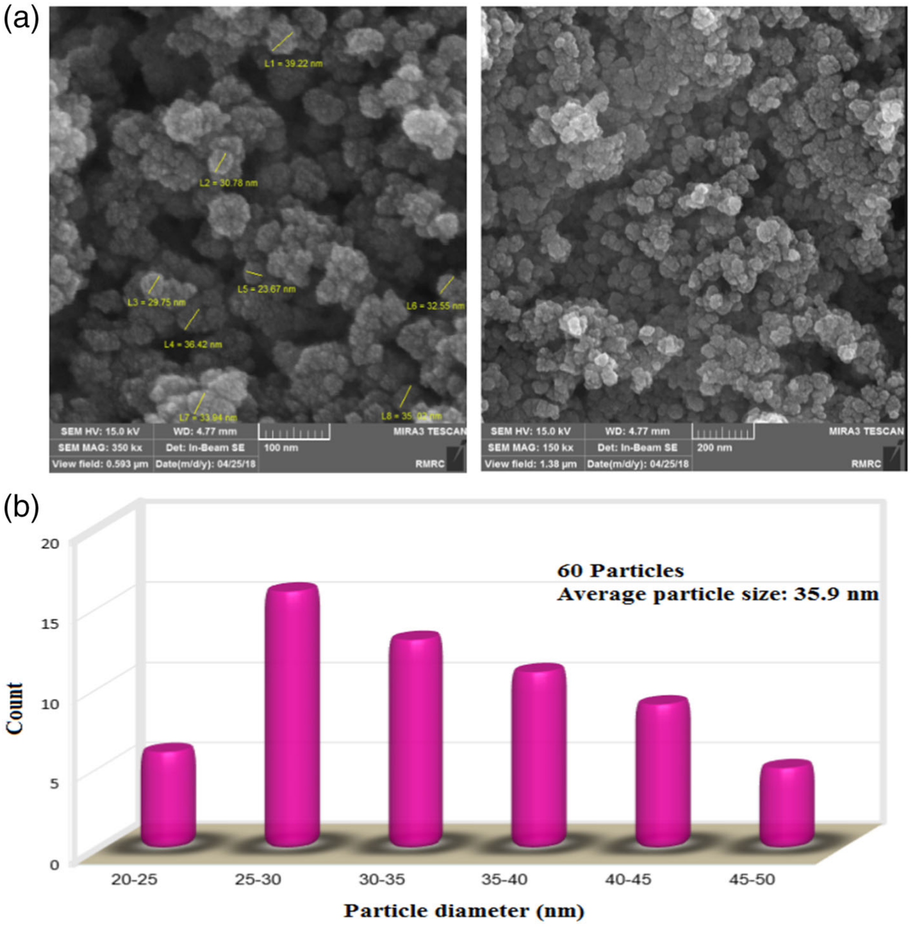


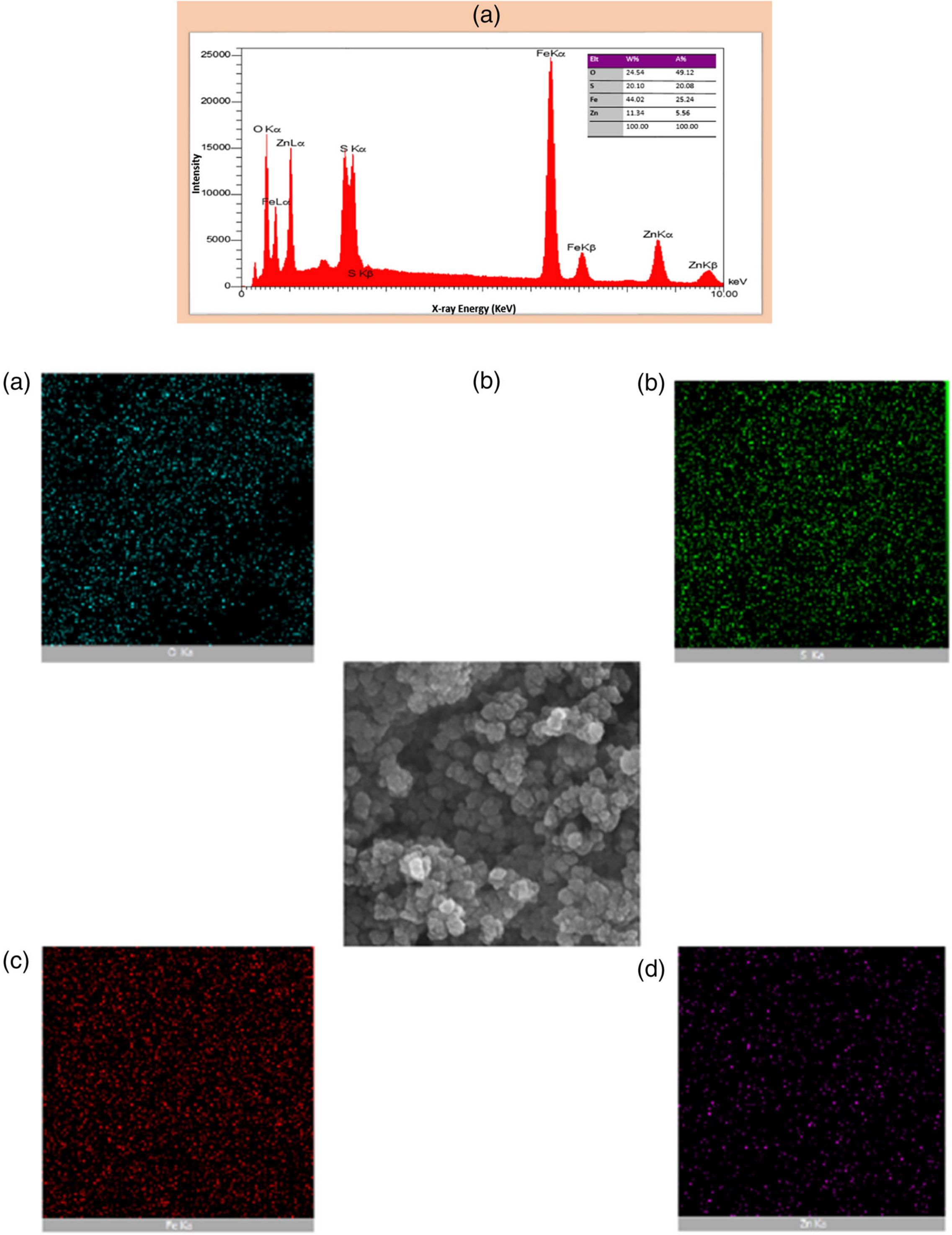
FIGURE 2 (a) SEM images and (b) particle size distribution diagram of the ZnS-$ZnFe_2O_4$ MTMO

acid that was characterized by VSM, FT-IR, SEM and EDS analysis. In fact, we examined the catalytic activity of the ZnS-$ZnFe_2O_4$ MTMO in 2,4,5-triaryl-1*H*-imidazoles. In this report, it was revealed that the catalytic efficiency of the ZnS-$ZnFe_2O_4$ for the MCRs of 2,4,5-triaryl-1*H*-imidazole using benzil **1**, aromatic aldehyde **2** and ammonium acetate **3** under ultrasonic irradiation in ethanol at 70 °C.

4 | CHARACTERIZATION OF THE ZNS-ZNFE$_2$O$_4$ HYBRID NANOCATALYST

4.1 | FT-IR spectra

FT-IR spectral analysis was used to examine and identify the functional groups. The results obtained in the



Elt	W%	A%
O	24.54	49.12
S	20.10	20.08
Fe	44.02	25.24
Zn	11.34	5.56
	100.00	100.00

FIGURE 3 (I) EDS diagram, and (II) elemental mapping images for (a) O, (b) S, (c) Fe and (d) Zn of the ZnS-$ZnFe_2O_4$ MTMO

FT-IR spectrum was used to verify the synthesis of the ZnS-$ZnFe_2O_4$ MTMO and to confirm the ZnS and $ZnFe_2O_4$ bond formations (Figure 1). According to the interactions between cation and oxygen in the tetrahedral and octahedral positions; in the spinel structure, two vibrational frequencies of $ZnFe_2O_4$ were shown at 665 cm^{-1}, 450 cm^{-1}. The absorption band at 665 cm^{-1} related to vibration of the tetrahedral position (Zn–O bond) and at 450 cm^{-1} is demonstrated with Fe^{3+} vibration frequencies at the octahedral positions (Fe–O bond).[32] The major absorption band of ZnS was observed at 966 cm^{-1}. As a result, they confirm stretching and bending absorption bands of the vibrations of the Zn-S. The intense absorption in the area between 3300–3700 cm^{-1} is related to the O-H stretching vibrations of water molecules, with its bending vibration in the area of 1627 cm^{-1}.[33] The absorption bands of 1350 and 1386 cm^{-1} relating to the interactions between $ZnFe_2O_4$ and ZnS in ZnS-$ZnFe_2O_4$.

4.2 | SEM analysis

SEM images of the synthesized nanocatalyst were shown in Figure 2a. The structure of the nanocatalyst was studied by this method and showed nearly spherical or Cauliflower-like morphology.[34] The approximately uniform particle size distribution was below 50 nm (Figure 2b).

4.3 | EDS analysis

Elemental composition of ZnS-$ZnFe_2O_4$ was identified by EDS analysis. As shown in Figure 3, iron, zinc, oxygen and sulfur are present elements in the nanocatalyst. In addition, the presence of all these elements in the recycled nanocatalyst confirmed its reusability.

4.4 | VSM magnetization curve

Magnetic properties of the ZnS-$ZnFe_2O_4$ MTMO were recorded by using a vibrating sample magnetometer curve (Figure 4). Due to the fact that the catalyst has a supramagnetic property that allows it to be separated from the solution by the use of external magnets. Magnetic saturations of ZnS-$ZnFe_2O_4$ and $ZnFe_2O_4$ were 52/9 and 66/9 electromagnetic units per gram (emu g^{-1}), respectively. In fact, the ZnS content in the ZnS-$ZnFe_2O_4$ structure has reduced its magnetic saturation value in comparison with $ZnFe_2O_4$. However, this MTMO with an external magnetic force can be easily separated from the reaction mixture.

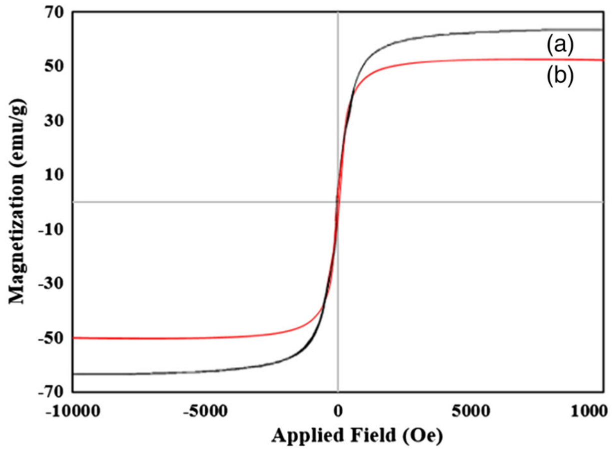


FIGURE 4 VSM curve of the (a) ZnS-$ZnFe_2O_4$ MTMO and (b) $ZnFe_2O_4$

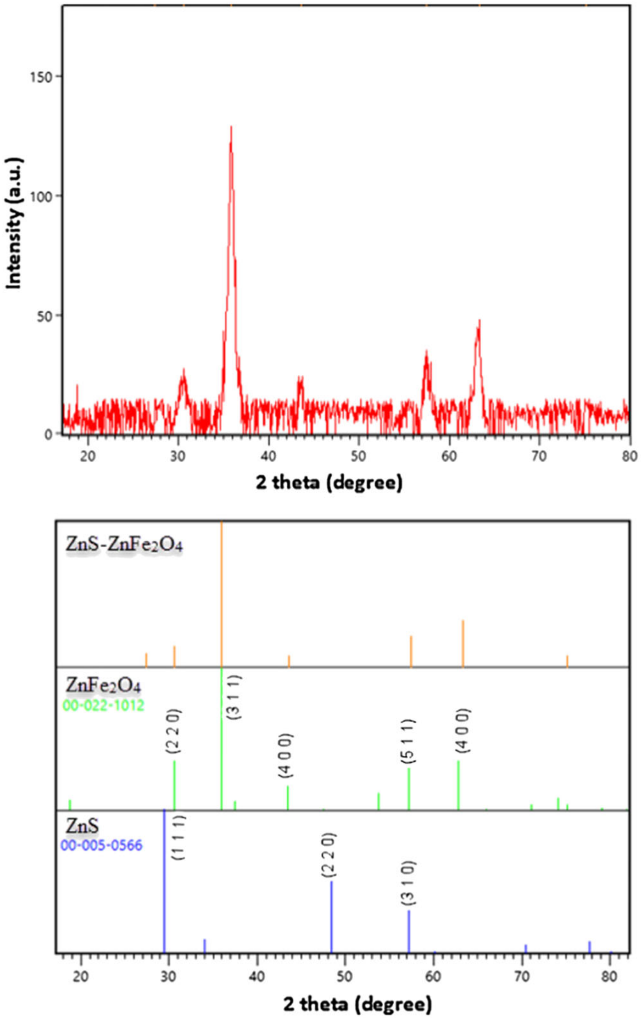


FIGURE 5 XRD pattern of the ZnS-$ZnFe_2O_4$ MTMO, ZnS and $ZnFe_2O_4$

4.5 | XRD pattern analysis

Figure 5 shows the XRD pattern of the ZnS-$ZnFe_2O_4$. In this figure, a comparison was made between the standard XRD patterns of $ZnFe_2O_4$, ZnS and the prepared nanocatalyst. As shown in the patterns, ZnS nanoparticles were confirmed with major peaks at $2\theta = 29, 48°$ and 57° by the standard diffraction card no. JCPDS, 05–0566. The XRD pattern of the $ZnFe_2O_4$ nanoparticles defined according to the standard diffraction card no. JCPDS, 22–1012 and major peaks at $2\theta = 30°, 36°, 43°$, 57° and 63°. ZnS and $ZnFe_2O_4$ have a cubic crystalline structure based on Xpert software. The XRD pattern of the ZnS-$ZnFe_2O_4$ MTMO indicates major peaks at $2\theta = 27°, 30°, 36°, 43°, 57°$ and 63° that confirms the presence of ZnS and $ZnFe_2O_4$ in the crystalline structure of the nanocatalyst.

4.6 | Application of the ZnS-$ZnFe_2O_4$ MTMO hybrid nanocatalyst for the synthesis of 2,4,5-triaryl-1*H*-imidazoles

ZnS-$ZnFe_2O_4$ MTMO was predictable to be more effective than ZnS and $ZnFe_2O_4$. In this regard and for a closer research on the application of this nanocatalyst, in a model reaction between benzil (1 mmol), 4-chlorobenzaldehyde (1 mmol) and ammonium acetate (4 mmol), the effect of the catalyst was studied (Table 2). To optimize the reaction, a mixture of aldehyde (1 mmol), benzil (1 mmol), ammonium acetate (4 mmol), and 2 mg of MTMO catalyst was sonicated at 70 °C for an appropriate time. To find the optimal conditions for the reaction, in the absence of any catalyst, the reaction yield was very low (entries 1–4). Maximum yield was gained (95%) when the reactions were used with 2 mg of the catalyst (entry 14). The higher amounts the catalyst loading did not lead to substantial change in the yield and time of the reaction. The reaction was investigated the effect of solvents such as EtOH, THF, CH_3CN and H_2O. But, none of the solvents were found to be more effective than ethanol. Thus, ethanol was used as an appropriate solvent for the reaction at 70 °C under ultrasonic irradiation (entries 13–16). A model reaction was also carried out with ZnS, $ZnFe_2O_4$ and ZnS-$ZnFe_2O_4$. These results approved that ZnS-$ZnFe_2O_4$ MTMO was more suitable for this reaction.

4.7 | Comparison of the catalyst activity

ZnS-$ZnFe_2O_4$ MTMO can be applied as an efficient nanocatalyst in MCRs. As shown in Table 3, ZnS-$ZnFe_2O_4$ showed superior activity in terms of time, temperature and even the efficiency in comparison with other reported catalysts such as Montmorilonite K10, Yb $(OPf)_3$ and Zr $(acac)_4$.

TABLE 2 Optimization of the model reaction conditions[a]

Entry	Catalyst	Catalyst loading (g)	Solvent	Temperature (°C)	Time (min)	Yield[b] (%)
1	-	-	Solvent-free	r.t.	290	-
2	-	-	EtOH	r.t.	270	Trace
3	-	-	EtOH	Ultrasonic/70	220	25
4	-	-	H_2O	Ultrasonic/70	250	Trace
5	ZnS	0.02	EtOH	Ultrasonic/70	155	45
6	$ZnFe_2O_4$	0.02	EtOH	Ultrasonic/70	140	40
7	ZnS-$ZnFe_2O_4$	0.02	EtOH	Ultrasonic/30	95	60
8	ZnS-$ZnFe_2O_4$	0.02	EtOH	Ultrasonic/50	80	75
9	ZnS-$ZnFe_2O_4$	0.02	EtOH	Reflux/80	160	30
10	ZnS-$ZnFe_2O_4$	0.03	H_2O	Ultrasonic/70	85	55
11	ZnS-$ZnFe_2O_4$	0.03	CH_3CN	Ultrasonic/70	120	46
12	ZnS-$ZnFe_2O_4$	0.03	THF	Ultrasonic/70	145	40
13	ZnS-$ZnFe_2O_4$	0.03	EtOH	Ultrasonic/70	50	68
14	ZnS-$ZnFe_2O_4$	0.02	EtOH	Ultrasonic/70	15	95
15	ZnS-$ZnFe_2O_4$	0.01	EtOH	Ultrasonic/70	70	65
16	ZnS-$ZnFe_2O_4$	0.005	EtOH	Ultrasonic/70	78	60

[a] Reaction conditions: 4-chlorobenzaldehyde (1 mmol), benzil (1 mmol), ammonium acetate (4 mmol), EtOH (1 ml) and ZnS-$ZnFe_2O_4$.

[b] Isolated yields.

TABLE 3 Comparison of various catalysts with ZnS-$ZnFe_2O_4$ MTMO in the synthesis of 2-(4-chlorophenyl)-4,5-diphenyl-1*H*-imidazole (**4d**)

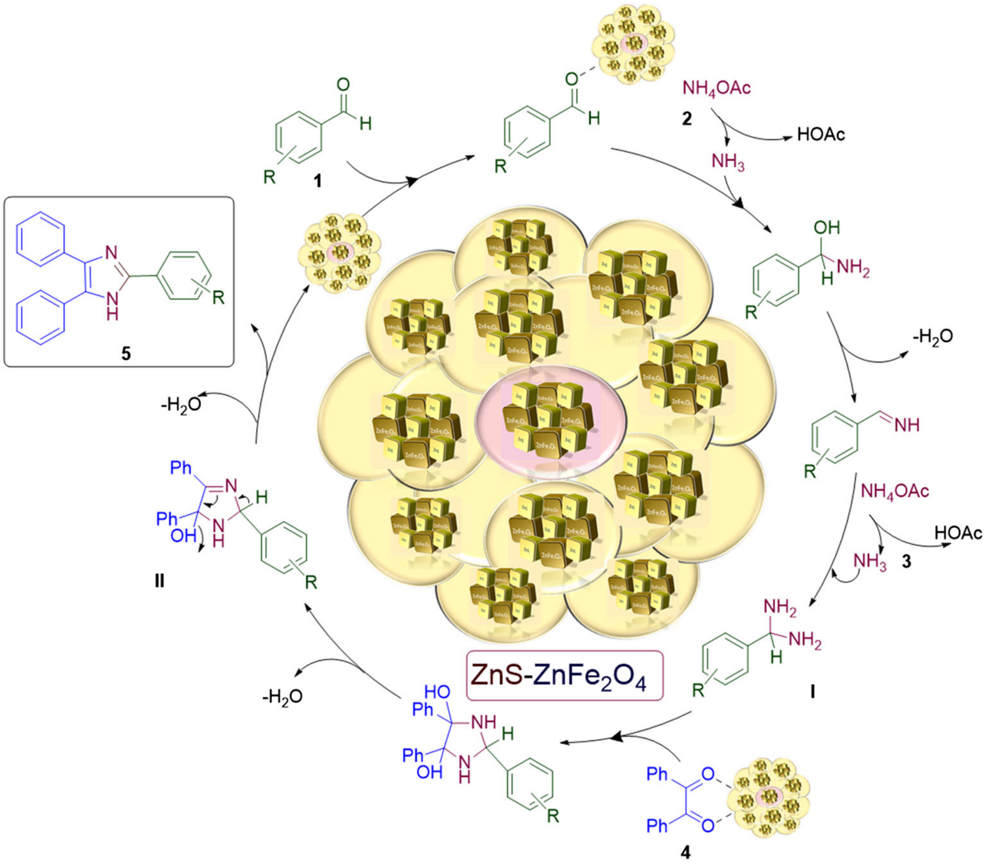
Entry	Catalyst	Solvent/ conditions	Time (min)	Yield (%)	[Lit.]
1	Montmorilonite K10	EtOH/reflux	95	75	[35]
2	Yb $(OPf)_3$	Perflurodecalin/ 80 °C	360	83	[36]
3	Zr $(acac)_4$	Ultrasonic/r.t.	35	91	[37]
4	ZnS-$ZnFe_2O_4$	Ultrasonic/ 70 °C	15	95	This work

4.8 | Proposed mechanism for the synthesis of 2,4,5-triaryl-1*H*-imidazoles in the presence of ZnS-$ZnFe_2O_4$ MTMO

The plausible mechanism for the synthesis of 2,4,5-triaryl-1*H*-imidazoles via MCR in the presence of ZnS-$ZnFe_2O_4$ MTMO is shown in Scheme 2. According to the mechanism, the ZnS-$ZnFe_2O_4$ MTMO as a Lewis acid, interacts with the oxygen of the carbonyl group of benzaldehyde. Then, ammonia as a nitrogen source arisen by ammonium acetate attacks to the activated carbonyl to form imine by removing an H_2O molecule. Next, intermediate **I** was formed from the nucleophilic attack of ammonia to imine. ZnS-$ZnFe_2O_4$ activates carbonyls of benzil to be reacted by **I** to result the formation of a cyclic intermediate **II**. Finally, by removing of an H_2O molecule and in order to reach the aromatic heterocyclic ring, the product of 2,4,5-triaryl-1*H*-imidazoles was provided.

4.9 | Reusability of the ZnS-$ZnFe_2O_4$ MTMO hybrid nanocatalyst

The recovered catalyst from the reaction mixture was reused six times for subsequent reactions without significant decrease of activity (Figure 6). As a result, a reusable, eco-friendly and efficient Lewis acid catalyst has been successfully used for one-pot synthesis of imidazole derivatives. In addition, the simplicity of the preparation and performance of the present nanocatalyst make this protocol as an effective and attractive approach for the future works and large scale industrial production.[38–45]



SCHEME 2 Proposed mechanism for the synthesis of 2,4,5-triaryl-1*H*-imidazoles by using ZnS-$ZnFe_2O_4$ MTMO.

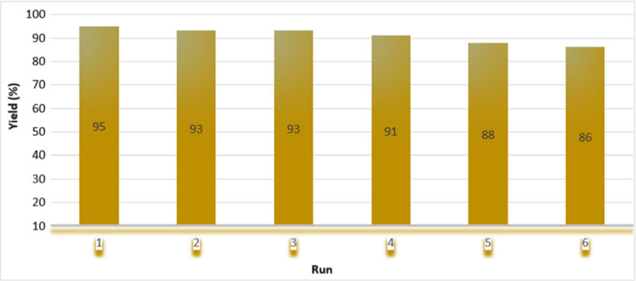


FIGURE 6 Reusability of the ZnS-$ZnFe_2O_4$ MTMO for the synthesis of **4d**

5 | CONCLUSIONS

In summary, in this work, the design, preparation, characterization and synergistic effect investigation of a novel bimetallic Lewis acid was described as a MTMO for the synthesis of 2,4,5-triaryl-1*H*-imidazoles. ZnS-$ZnFe_2O_4$ MTMO as an efficient heterogeneous magnetic MTMO was successfully synthesized via the co-precipitation method through a facial procedure. Then, ZnS-$ZnFe_2O_4$ MTMO was used for the one-pot MCR of benzil, various aldehydes and ammonium acetate in ethanol under ultrasonic irradiation (Scheme 1) in excellent yields and short reaction times and easy performance. The advantages of this approach include mild reaction conditions, high product yields and easy work-up procedure, simple recyclability, high atom economy and eco-friendly aspects of the present heterogeneous nanocatalyst. According to SEM images (Figure 2a), morphology of this MTMO was almost spherical and Cauliflower-like with average particle size of about 36 nm (Figure 2b) and magnetic saturation of ZnS-$ZnFe_2O_4$ and $ZnFe_2O_4$ were 52.9 and 66.9 emu g^{-1}, respectively (Figure 4). As a result, those confirmed successful preparation of the new hybrid MTMO as well as its efficiency in MCRs. In addition, due to high potential of this newly introduced MTMO, it can be developed to diverse further applications in a wide variety of investigations.

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CONFLICT OF INTEREST

All authors express that they have no conflict of interest.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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