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Synthesis of mono- and bis-spiro-2-amino-4*H*-pyrans catalyzed by *S*-alkyl *O*-hydrogen sulfothioate functionalized silica-coated magnetic nanoparticles under ultrasound irradiation

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Abstract New *S*-alkyl *O*-hydrogen sulfothioate functionalized silica-coated magnetic nanoparticles were prepared in a simple method and evaluated as an efficient catalyst in synthesis of mono- and bis-spiro-2-amino-4*H*-pyrans through three or pseudo-five component reaction of isatins or bis-isatins and malononitrile with dimedone or 4-hydroxycoumarin. The yields were excellent, and the catalyst could be recovered simply from the reaction mixture using an external magnet and reused at least five times without noticeable loss of catalytic activity.

Keywords Magnetic nanoparticles · Mono- and bis-spiro-2-amino-4*H*-pyrans · Ultrasound irradiation · 1,3-Dicarbonyl compounds

Introduction

Among the heterocyclic compounds, 2-amino-4*H*-pyrans have received considerable attention due to a wide range of biological properties such as antiallergic [1], antitumor [2] and antibacterial activities [3]. They have been used in pigments and cosmetics and as potentially biodegradable agrochemicals [1, 4]. Spiropyrans are used in photochromic compounds, and they have been widely studied because of their potential utility in various high-tech applications [5]. Spirooxindole derivatives display various bioactive effects such as antitumor and antimicrobial, and as inhibitors of the human NKI receptor [6]. The existence of spiroindoline derivatives in the structure of organic compounds could potentially have novel therapeutic activities and thus there is an increased interest in obtaining spiro-2-amino-4*H*-pyrans containing oxindole moiety [7–21].

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Recently, spiro-2-amino-4*H*-pyrans were synthesized by multi-component condensation reaction of cyclic ketone such as isatin with malononitrile and β-dicarbonyl compounds such as 4-hydroxycoumarin, dimedone or barbituric acid. This condensation has been catalyzed by a variety of reagents, such as ammonium salt [7], triethylbenzylammonium chloride (TEBA) [8], [BMIm]BF_4 [9], nano-MgO [10], $InCl_3/SiO_2$ [11], (SB-DBU)Cl [12], alum [13], sodium stearate [14], SBA-Pr-NH_2 [15], L-proline [16], magnesium perchlorate [17], ethylenediamine diacetate [18, 19], mesoporous silica nanoparticles [20], and sulfuric acid-modified poly(ethylene glycol) [21]. Also, this condensation has been performed in an electrocatalytic procedure [22]. Although each of the methods has their own merits, some methods suffer from long reaction times, low yields, harsh reaction conditions, and tedious workup. The recovery of the catalysts is an important problem. Therefore, the development of alternative methods for the synthesis of these valuable compounds could be very worthwhile.

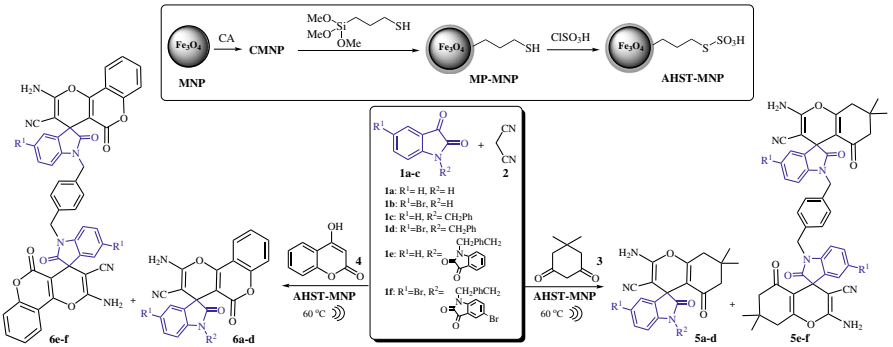
The use of acid-functionalized silica-coated magnetic nanoparticles has received considerable attention in organic synthesis due to their environmental compatibility; ease of handling, non-toxic nature, their reusability, and facile separation from the products [23–29]. Various acid-functionalized silica-coated magnetic nanoparticles have been prepared and used as efficient catalysts for different organic transformations, such as deprotection of benzaldehyde dimethylacetals [23], synthesis of α-amino nitriles [24], synthesis of mono-, di- and tri-[bis(6-aminopyrimidonyl) methanes] [25], quinolones [26], synthesis of benzo[α]xanthenone derivatives [27], aminoimidazopyridine skeletons [28], and 3,4-dihydropyrimidinones/thiones [29].

Although three-component reaction of dimedone with isatin has been investigated, there is no report on the reaction of dimedone with bis-isatins for the synthesis of bis-spiro-2-amino-4*H*-pyrans. As a continuation of our research into the preparation and catalytic investigation of new acid-functionalized silica-coated magnetic nanoparticles [25], and due to the biological activity of spiro-2-amino-4*H*-pyrans, we prepared new *S*-alkyl *O*-hydrogen sulfothioate functionalized silica-coated magnetic nanoparticles (AHST-MNPs) by direct reaction of chlorosulfonic acid with mercaptopropyl silica-coated magnetic nanoparticles (MPS-MNPs), and it was successfully used as a catalyst in the synthesis of mono- and bis-spiro-2-amino-4*H*-pyrans under ultrasonic irradiation at 60 °C (Scheme 1).

Experimental

General

Isatin **1**, malononitrile **2**, dimedon **3**, 4-hydroxycoumarin **4**, $FeCl_3$, 3-(trimethoxysily)-1-propanthiol (TMSPT), and solvents were used without further purification. Fe_3O_4 nanoparticles (MNP) and citric acid-modified nanoparticles (CMNPs) were prepared according to the literature procedure [30]. Sonication was performed in a Struers Metason 200 HT ultrasonic cleaner with a frequency of 50–60 Hz and an output power of 140 W. The products were characterized by elemental analysis and by IR, ^{1}H NMR, and ^{13}C NMR spectroscopy. Fourier-transform IR spectra were



Scheme 1 Preparation of *S*-alkyl *O*-hydrogen sulfothioate functionalized silica-coated magnetic nanoparticles (AHST-MNP) and synthesis of mono- and bis-spiro-2-amino-4*H*-pyrans catalyzed by AHST-MNP

recorded by using a Unicom Galaxy Series FTIR 5000 Spectrophotometer. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker Avance 400 MHz spectrometer at 400 and 100 MHz, respectively. Elemental analyses were performed by using Vario EL III elemental analyzer. Check spectroscopy, X-ray diffraction (XRD) by X″ 3064 Pert Pw done. The size and shape of nanoparticles by scanning electron microscope (SEM; Hitachi S-4160) were identified as devices. TGA data samples by TG Perkin-Elmer 6300 model PYRIS TM Diamond were recorded.

General procedure for the preparation of mercaptopropyl silica-coated magnetic nanoparticles (MPS-MNPs)

An amount of 1.5 g of CMNPs was dispersed in 250 mL ethanol:water (1:1) mixture and sonication was carried out for 30 min to maintain proper dispersion. Under continuous mechanical stirring, 2.5 mL of TMSPT was added. After mechanical agitation at 40 °C for 4 h, the black precipitate was isolated by magnetic decantation and washed with de-ionized water and ethanol, and then dried at room temperature.

General procedure for the preparation of S-alkyl O-hydrogen sulfothioate functionalized silica-coated magnetic nanoparticles (AHST-MNPs)

The prepared MPS-MNPs were added to a two-necked flask fitted with a constant-pressure dropping funnel and a tube to remove the HCl gas formed, by conducting it to an adsorbent solution. Chlorosulfonic acid (1.5 mL) was added dropwise over a period of 30 min at room temperature and followed by slow mechanical stirring. HCl gas immediately evolved from the reaction vessel. After that, the mixture was shaken well for 30 min. The AHST-MNPs were washed with acetone and distilled water to remove excess chlorosulfonic acid and finally dried in an oven at 60 °C for 6 h.

General procedure for the preparation of mono- and bis-spiro-2-amino-4H-pyrans ***5a–f*** *and* ***6a–f***

A mixture of isatin **1** or **2** (1 mmol), malononitrile (1 mmol for **1** and 2 mmol for **2a**, **b**), dimedone or 4-hydroxycoumarin (1 mmol for **1a**, **b** and 2 mmol for **2a**, **b**) and AHST-MNPs (0.07 g) in H_2O or EtOH/H_2O (4:1) (10 mL) was exposed to US irradiation at 60 °C for the appropriate time (see Table 2, below). Upon completion of the reaction as monitored by TLC, the AHST-MNPs were removed under applied external magnetic field. Next, the solution was reduced in volume and left for slow evaporation. The solid was collected by filtration and washed with H_2O (15 mL) and cold EtOH (15 mL) to give the desired products. The crude products were recrystallized from EtOH.

*Spiro-2-amino-4H-pyrans (**5a**)*

IR (KBr) (ν_{max}, cm^{-1}): 3,302, 3,136, 2,960, 2,928, 2,193, 1,722, 1,649, 1,604, 1,471, 1,348, 1,055, 746. ^{1}H-NMR (DMSO-d_6, 400 MHz) δ_H: 10.40 (1H, s, NH), 7.23 (2H, s, NH_2), 7.12–7.16 (1H, t, $J = 7.2$ Hz, H_{arom}), 6.97–6.99 (1H, d, $J = 7.2$ Hz, H_{arom}), 6.87–6.91(1H, t, $J = 7.2$ Hz, H_{arom}), 6.78–6.80 (1H, d, $J = 7.6$ Hz, H_{arom}), 2.50–2.57 (2H, ABq, CH_2), 2.12–2.16 (2H, ABq, CH_2), 1.04 (3H, s, CH_3), 1.00 (3H, s, CH_3).

*Spiro-2-amino-4H-pyrans (**5b**)*

IR (KBr) (ν_{max}, cm^{-1}): 3,311, 3,149, 2,958, 2,928, 2,195, 1,724, 1,651, 1,604, 1,475, 1,348, 1,057, 810, 667. ^{1}H-NMR (DMSO-d_6, 400 MHz) δ_H: 10.56 (1H, s, NH), 7.33 (2H, s, NH_2), 7.30 (1H, s, H_{arom}), 7.21(1H, s, H_{arom}), 6.75–6.77 (1H, d, $J = 8$ Hz, H_{arom}), 2.50–2.58 (2H, ABq, CH_2), 2.12–2.16 (2H, ABq, CH_2), 1.03 (6H, s, CH_3). Anal. calc. for $C_{19}H_{16}BrN_3O_3$: C, 55.09; H, 3.89; Br, 19.29; N, 10.14 %. Found: C, 55.20; H, 3.80; Br, 19.33; N, 10.22 %.

*Spiro-2-amino-4H-pyrans (**5c**)*

IR (KBr) (ν_{max}, cm^{-1}): 3,381, 3,319, 3,205, 2,964, 2,197, 1,712, 1,662, 1,601, 1,467, 1,352, 1,051, 893. ^{1}H-NMR (DMSO-d_6, 400 MHz) δ_H: 7.48–7.50 (2H, d, $J = 7.2$ Hz, H_{arom}), 7.26–7.33 (4H, m, NH_2, H_{arom}), 7.08–7.13 (3H, m, H_{arom}), 6.94–6.98 (1H, t, $J = 7.6$ Hz, H_{arom}), 6.68–6.70 (1H, d, $J = 7.6$ Hz, H_{arom}), 4.86–4.96 (2H, ABq, CH_2), 2.50–2.62 (2H, ABq, CH_2), 2.15–2.24 (2H, ABq, CH_2), 1.06 (3H, s, CH_3), 1.02 (3H, s, CH_3).

*Spiro-2-amino-4H-pyrans (**5d**)*

IR (KBr) (ν_{max}, cm^{-1}): 3,317, 3,252, 3,176, 2,958, 2,197, 1,710, 1,674, 1,608, 1,467, 1,352, 1,053,748, 669. ^{1}H-NMR (DMSO-d_6, 400 MHz) δ_H: 7.47 (1H, s, H_{arom}), 7.43–7.46 (1H, d, $J = 9.6$ Hz, H_{arom}), 7.25–7.35 (7H, m, NH_2, H_{arom}),

6.66–6.68 (1H, d, $J = 8.8$ Hz, H_{arom}), 4.86–4.95 (2H, ABq, CH_2), 2.50–2.66 (2H, ABq, CH_2), 2.20–2.24 (2H, t, $J = 16.8$ CH_2), 1.04 (6H, s, CH_3).

*Bis-spiro-2-amino-4H-pyrans (**5e**)*

IR (KBr) (ν_{max}, cm^{-1}): 3,393, 3,292, 3,167, 2,958, 2,197, 1,710, 1,674, 1,608, 1,467, 1,352, 1,053, 748. ^{1}H-NMR (DMSO-d_6, 400 MHz) δ_H: 7.34 (4H, s, NH_2), 7.24 (2H, s, H_{arom}), 6.98–7.05 (6H, m, H_{arom}), 6.85–6.89 (2H, t, $J = 15.2$ Hz, H_{arom}), 6.58–6.60 (2H, d, $J = 7.6$, H_{arom}) 4.79 (4H, s, CH_2), 2.41–2.52 (4H, m, CH_2), 2.05–2.11 (4H, ABq, CH_2), 0.96 (6H, s, CH_3), 0.93 (6H, s, CH_3). ^{13}C-NMR (DMSO-d_6, 100 MHz) δ_C: 26.98, 27.59, 31.96, 43.06, 49.84, 58.40, 70.58, 108.91, 109.94, 110.59, 117.34, 122.53, 122.89, 127.09, 128.26, 133.54, 134.99, 142.54, 158.87, 164.49, 176.68, 195.04.

*Bis-spiro-2-amino-4H-pyrans (**5f**)*

IR (KBr) (ν_{max},cm^{-1}): 3,433, 3,393, 2,975, 2,474, 2,193, 1,709, 1,674, 1,639, 1,481, 1,352, 1,101, 808, 617. ^{1}H-NMR (DMSO-d_6, 400 MHz) δ_H: 7.34 (4H, s, NH_2), 7.32 (2H, s, H_{arom}), 7.22–7.25 (6H, m, H_{arom}), 6.56–6.58 (2H, d, $J = 9.2$ Hz, H_{arom}), 4.98 (4H, s, CH_2), 2.44–2.53 (4H, m, CH_2), 2.03–2.10 (4H, t, $J = 28.4$ Hz, CH_2), 0.93 (12H, s, CH_3). ^{13}C-NMR (DMSO-d_6, 100 MHz) δ_C: 27.15, 27.51, 32.05, 43.46, 49.79, 67.99, 81.51, 103.64, 109.67, 110.89, 114.40, 117.25, 117.82, 127.10, 131.01, 134.70, 135.90, 140.33, 140.96, 158.99, 176.34, 195.32.

Results and discussion

The activity of the AHST-MNP catalyst was evaluated for the synthesis of mono- and bis-spiro-2-amino-4*H*-pyrans. AHST-MNP behaves as an organic/inorganic hybrid (interphase) in which a Brønsted acid site has been created. The catalyst is isolated easily with a magnet and finally the high surface area of the AHST-MNP means that they can accommodate large amounts of the catalytic species, thus making the catalyst highly active. In order to optimize the conditions, the reactions were carried out under different conditions. The condensation of isatin **1a**, malononitrile **2** and dimedone **3** was chosen as a model.

The effect of the amount of catalyst on the yield was investigated (Table 1, entries 1–4). It was found that 0.07 g of catalyst for 1 mmol of isatin was the appropriate amount for the reaction (Table 1, entry 3). The solvents also played an important role in this reaction catalyzed by AHST-MNPs. The use of EtOH:H_2O, DMF, THF and DMSO as solvents furnished poor yields (Table 1, entries 6–9). The reaction hardly proceeded in THF. However, the reaction in water afforded the product in high yield with nearly complete conversion. Therefore, water was selected as the reaction solvent in the following investigation (Table 1, entry 3). Hence, the best reaction conditions were obtained by using 0.07 g of AHST-MNP in water at 60 °C.

Table 1 Optimization of reaction for the synthesis of **5a** under ultrasonic irradiation at 60 °C

Entry[a]	AHST-MNPs	Solvent	Yield
1	0.03	H_2O	64
2	0.05	H_2O	85
3	0.07	H_2O	94
4	0.10	H_2O	94
5	–	H_2O	28
6	0.07	EtOH:H_2O	67
7	0.07	DMF	58
8	0.07	THF	20
9	0.07	DMSO	40

[a] Isatin (1 mmol), malononotrile (1 mmol), dimedone (1 mmol). Time: 105 min, temp.: 60 °C

Mono-spiro-2-amino-4*H*-pyrans **5a–d** and **6a–d** were obtained by the three-component condensation of 1 mmol of isatin **1a–c** and 1 mmol malononitrile **2** and 1 mmol dimedone **3** or 4-hydroxycoumarin **4** in the presence of 0.07 gr AHST-MNP as heterogeneous catalyst. The reaction was carried out in water (10 mL) at 60 °C under ultrasonic conditions to give products **5a**, **b** and **6a–d** in high yields. Bis-spiro-2-amino-4*H*-pyrans **5e**, **f** and **6e**, **f** were achieved by pseudo five-component reaction of **1d**, **e** (1 mmol) with malononitrile **2** (2.1 mmol) and dimedone or 4-hydroxycoumarin (2.1 mmol) in the presence of 0.07 g AHST-MNP in water (10 mL) under ultrasonic irradiation at 60 °C. We found that the catalyst AHST-MNP showed high catalytic activity and could be recovered and recycled several times without significant loss of activity (Table 2).

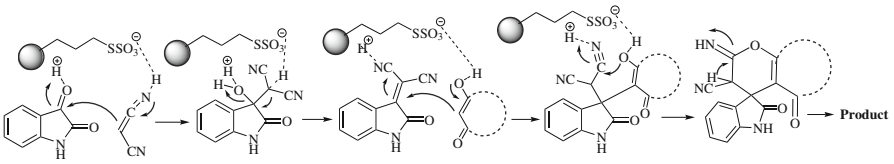
AHST-MNPs were obtained in a simple one-step procedure with direct reaction of chlorosulfonic acid with mercaptopropyl silica-coated magnetic nanoparticles. For the synthesis of mercaptopropyl silica-coated magnetic nanoparticles, prior to functionalizing the magnetic particles with chlorosulfonic acid, modification of the nanoparticles' surface with negatively charged citrate groups was carried out and then the surface of these particles (CMNPs) was coated with mercaptopropyl trimethoxy silane to afford mercaptopropyl silica-coated magnetic nanoparticles.

The prepared catalyst (AHST-MNP) was characterized with IR, XRD, TGA and SEM. In FT-IR spectra, a visible spectrum is absorbed Fe–O bond stretching has appeared at 559 cm^{-1}. The peaks at 1,003 and 1,170 cm^{-1} correspond to the Si–O stretch and the broad band at 3,000–3,500 cm^{-1} and peak at 2,931 are due to $–SO_3H$ and –CH stretching vibrations, respectively.

Although we have not yet established the mechanism, a possible explanation is given in Scheme 2.

The sizes of nanoparticles were determined by scanning electron microscopy (SEM) (Fig. 1). SEM images of AHST-MNP are shown in Fig. 1. The SEM photograph illustrated that the AHST-MNP is spherical in shape and the average size of AHST-MNP is about 28 nm.

The thermal properties of AHST-MNP were analyzed by thermal gravimetric analysis (TGA) in the temperature range 40–850 °C under a nitrogen atmosphere.



Scheme 2 Plausible mechanism of the AHST-MNP catalyzed synthesis of 2-amino-4*H*-pyrans

Table 2 AHST-MNP catalyzed synthesis of mono- and bis-spiro-2-amino-4*H*-pyrans at 60 °C under ultrasonic irradiation

Entry	Isatin	Product	Time (h)/yield (%)	M_p (°C)	M_p (°C)$^{Lit.}$
1[a]	**1a**	Spiro-2-amino-4*H*-pyran (**5a**)	1:45/94	305	292 [9]
2[a]	**1b**	Spiro-2-amino-4*H*-pyran (**5b**)	1:45/92	313	
3[a]	**1c**	Spiro-2-amino-4*H*-pyran (**5c**)	2:15/91	275	268 [9]
4[a]	**1d**	Spiro-2-amino-4*H*-pyran (**5d**)	2:15/89	300	
5[b]	**1e**	Bis-spiro-2-amino-4*H*-pyran (**5e**)	4/77	>300	
6[b]	**1f**	Bis-spiro-2-amino-4*H*-pyran (**5f**)	4/79	>300	
7[a]	**1a**	Spiro-2-amino-4*H*-pyran (**6a**)	2.5/90	299–301	292–294 [7]
8[a]	**1b**	Spiro-2-amino-4*H*-pyran (**6b**)	2.5/89	>300	>300 [7]
9[a]	**1c**	Spiro-2-amino-4*H*-pyran (**6c**)	2.5/91	275–277	277–279 [7]
10[a]	**1d**	Spiro-2-amino-4*H*-pyran (**6d**)	2.5/88	>300	
11[b]	**1e**	Bis-spiro-2-amino-4*H*-pyran (**6e**)	4/76	>300	318 [13]
12[b]	**1f**	Bis-spiro-2-amino-4*H*-pyran (**6f**)	4/77	268–271	268 [13]

[a] Solvent: H_2O

[b] Solvent: EtOH/H_2O (4:1)

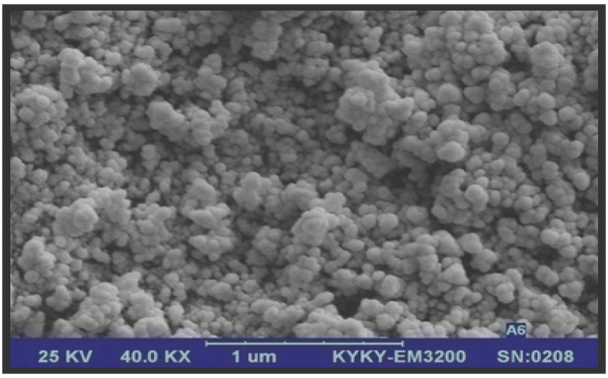


Fig. 1 SEM image of AHST-MNP

The primary weight loss up to 160 °C was related to the removal of physically adsorbed solvent. The rate of weight loss between 160 and 630 °C is relatively slow representing the high thermal stability of the AHST-MNP, which is generally

thermally stable until 630 °C, and the maximum rate of lost weight for these nanoparticles was started from 540 °C. There is a well-defined mass weight loss of 44 % between 160 and 700 °C related to the breakdown of the sulfuric acid and alkyl thiol moieties. In XRD patterns of the AHST-MNP, the intensity of the 43.61 reflection of the AHST-MNP was decreased after the introduction of the sulfonic acid (SA) group.

Conclusions

In conclusion, we have described an efficient and environmentally benign method for the preparation of mono- and bis-spiro-2-amino-4*H*-pyrans. This multi-component condensation reaction is efficiently catalyzed by AHST-MNPs in water at 60 °C under ultrasound irradiation. Operational simplicity, mild reaction conditions, enhanced rates, and high isolated yields of the pure products are significant advantages of the protocol presented here. AHST-MNP are the most promising catalyst because of their ease of handling and ease of recovery with external magnetic field.

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