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$[Fe_3O_4@SiO_2@Si(CH_2)_3–NMe_2–(CH_2)_2–NMe_2–SO_3H][Cl]_2$ as a novel magnetic nanocatalyst for the synthesis of bis(6-amino-1,3-dimethyl-uracil-5-yl)methanes

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Original Research

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Abstract:

A new magnetic nanomaterial, namely $[Fe_3O_4@SiO_2@Si(CH_2)_3–NMe_2–(CH_2)_2–NMe_2–SO_3H][Cl]_2$ (FSCNSC) was synthesized, and characterized by energy-dispersive X-ray spectroscopy (EDS), elemental mapping, field emission scanning electron microscopy (FE-SEM), X-ray diffraction (XRD), FT-IR, vibrating-sample magnetometery (VSM), thermogravimetric (TG) and derivative thermogravimetry (DTG) analyses. Afterward, it was used as a highly effective catalyst for the synthesis of bis(6-amino-1,3-dimethyluracil-5-yl)methanes under solvent-free conditions. FSCNSC was better than many of the reported catalysts for the synthesis of this category of uracil derivatives in terms of the reaction conditions, the reaction times, and yields.

Keywords: $[Fe_3O_4@SiO_2@Si(CH_2)_3–NMe_2–(CH_2)_2–NMe_2–SO_3H][Cl]_2$; Magnetic nanocatalyst; bis(6-amino-1,3-dimethyluracil-5-yl)methanes; 6-Amino-1,3-dimethyluracil; Solvent-free

1. Introduction

Currently, an attractive and useful research field is the production of magnetic nanomaterials and their application for diverse purposes since these materials have several unbeatable characteristics, including suitable thermal and chemical durability, efficacy, capability for functionalization, magnetically separable from process medium and high surface-to-volume ratio [1–29]. Magnetic nanomaterials have been applied as effective heterogeneous catalysts in organic synthesis [1–21]. They have been also used in supercapacitors [22] and fuel cells [23]. Furthermore, these substances have been utilized for CO_2 absorption [24], oil/water separation [25], determination of 6-tioguanine [26], extracting and preconcentration of pesticides from fruit juice [27], photodegradation of dyes [28] and degradation of antibiotics [29].

"Solvent-free conditions" is a practical, green, useful and highly efficient protocol in organic synthesis, and its advantages have been mentioned in the literature [30–35]. Uracil derivatives are essential components in the structure of RNA and have vital tasks in human life [36]. They also have many biological and pharmaceutical properties, e.g., antifungal [37], antibacterial [37], carbonic anhydrase inhibitor [38], antiviral [39], thymidine phosphorylase inhibitor [40], antiproliferative [41], antioxidant [42] and antitumor [43] activities. Furthermore, 6-aminouracils and their derivatives have been applied as scaffolds for the construction of some biologically active compounds, such as penciclovir, caffeine, theophylline, and theobromine [44–46]. The reaction of 6-amino-1,3-dimethyluracil (2 eq.) with aryl aldehydes (1 eq.) in the presence of a catalyst, leads to a significant category of uracil derivatives, namely bis(6-amino-1,3-dimethyluracil-5-yl)methanes [47–53]. It is worth noting that a few catalysts have been reported for this reaction; additionally, the reported methods are associated with one or more of these drawbacks: moderate yields, long reaction times, utilization of toxic organic solvent as the reaction medium, and need to special apparatuses (ultrasonic and microwave ovens). Thus, introducing new catalysts to promote the fabrication of bis(6-amino-1,3-dimethyluracil-5-yl)methanes is highly desirable.

Due to high importance of the uracil derivatives and the advantages of magnetic nanocatalysts, we have constructed a novel acidic catalyst supported on magnetite nanoparticles namely [Fe_3O_4 @SiO_2 @$Si(CH_2)_3$–NMe_2–$(CH_2)_2$–NMe_2–SO_3H][Cl]$_2$ (FSCNSC), and characterized it using various analyses such as EDS, elemental mapping, FE-SEM, XRD, FT-IR, VSM, TG, and DTG. Then, we used the nanomaterial to catalyze the fabrication of bis(6-amino-1,3-dimethyluracil-5-yl)methanes through the reaction of 6-amino-1,3-dimethyluracil (2 eq.) with aryl aldehydes (1 eq.) under solvent-free conditions. It is noteworthy that FSCNSC has none of the above-mentioned disadvantages.

2. Experimental

2.1 Chemicals and devices

The details of the materials and devices used have been given in supplementary material.

2.2 Procedure for the production of fscnsc (Scheme 1)

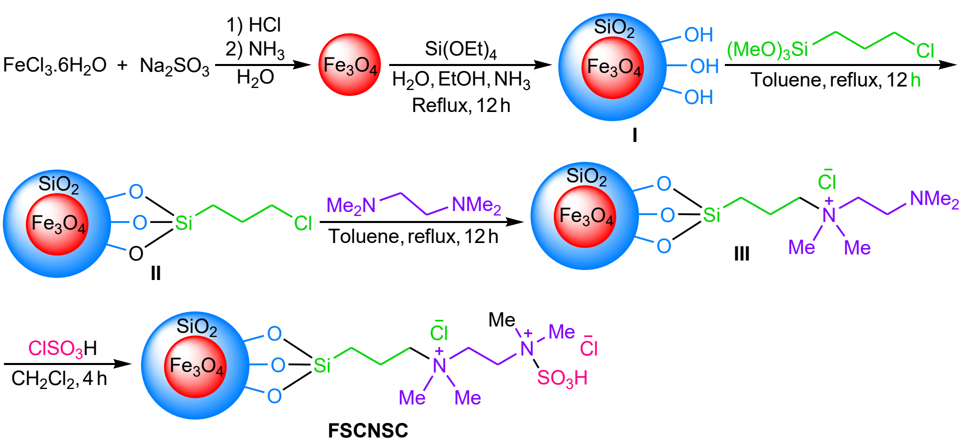
A reported procedure was used for the preparation of Fe_3O_4 [54, 55]. Afterward, a mixture of Fe_3O_4 (1.16 g, 5 mmol), $Si(OEt)_4$ (3.5 mL), EtOH (93 mL), H_2O (23 mL) and ammonia (3.5 mL) was stirred under reflux conditions for 12 h; the reaction mixture was cooled to ambient temperature, and the produced precipitate was magnetically separated, washed by $EtOH/H_2O$ (4/1), and dried to produce **I** [55, 56]. A mixture of (3-chloropropyl) trimethoxysilane (0.92 g, 5 mmol) and compound **I** (all the amount produced in the previous step) in dry toluene (40 mL) was stirred and refluxed under a flow of nitrogen gas for 12 h; the mixture was cooled to room temperature, and the solid was isolated by a magnet, washed by toluene (5 mL) and dried to construct **II** [55, 56]. Thereafter, *N, N,N′,N′*-tetramethylethylenediamine (0.75 mL, 5 mmol) and toluene (30 mL) were added to compound **II** (all the amount produced in the previous step), and the resulting mixture was stirred and refluxed for 12 h; after cooling the mixture to room temperature, the resulting precipitate was magnetically isolated, washed by toluene (5 mL), and dried to afford **III**. Finally, **III** (all the amount produced in the previous step) was added gradually to a stirring solution of $ClSO_3H$ (0.34 mL, 5 mmol) in dry CH_2Cl_2 (25 mL) at 10 °C, and the mixture was stirred for 4 h at room temperature; the produced solid was isolated by a magnet, washed by CH_2Cl_2 (2×5 mL), and dried to fabricate FSCNSC.

Note: when the reactants were mixed (for the construction of **I**, **II**, **III** and FSCNSC), initially, the resulting mixtures were sonicated at room temperature for 20 min, and then, the reactions were performed.

2.3 General procedure for the synthesis of bis(6-amino-1,3-dimethyluracil-5-yl)methanes

A mixture of 6-amino-1,3-dimethyluracil (0.31 g, 2 mmol), aldehyde (1.05 mmol), and FSCNSC (0.06 g) was vigorously stirred by a rod at 120 °C for the necessary time. After the completion of the reaction was approved by TLC, the mixture was cooled to room temperature, EtOAc (30 mL) was added, stirred for 2 min under reflux conditions, and the catalyst was magnetically isolated. The solvent was distilled, and the resulting precipitate was recrystallized from ethanol (95%) to afford the pure products **(3)**.

The ^1H and ^{13}C NMR spectra of some bis(6-amino-1,3-dimethyluracil-5-yl)methanes **(3)** have been given in supplementary material.



Scheme 1. the production of FSCNSC.

2.4 Selected spectral data of the synthesized bis(6-amino-1,3-dimethyluracil-5-yl)methanes

*Compound **3b***

^{1}H NMR (300 MHz, DMSO-d_6): δ (ppm): 3.07 (s, 3H, N–CH_3), 3.21 (s, 3H, N–CH_3), 3.37 (s, 6H, 2N–CH_3), 5.49 (s, 1H, $CH_{methine}$), 6.98 (broad, 2H, NH_2), 7.09 (dt, J = 7.5, 1.7 Hz, 1H, H_{Ar}), 7.27 (dt, J = 7.7, 1.4 Hz, 1H, H_{Ar}), 7.38 (dd, J = 7.8, 1.7 Hz, 1H, H_{Ar}), 7.48 (dd, J = 7.8, 1.4 Hz, 1H, H_{Ar}), 7.54 (broad, 2H, NH_2); ^{13}C NMR (75 MHz, DMSO-d_6): δ (ppm): 28.1, 28.5, 30.5, 37.3, 85.7, 86.9, 123.3, 127.4, 127.9, 129.8, 131.5, 133.3, 140.4, 150.9, 154.6, 162.4, 163.4.

*Compound **3c***

^{1}H NMR (500 MHz, DMSO-d_6): δ (ppm): 3.13 (s, 6H, 2N–CH_3), 3.31 (s, 3H, N–CH_3), 3.33 (s, 3H, N–CH_3), 5.54 (s, 1H, $CH_{methine}$), 7.10 (d, J = 8.1 Hz, 2H, H_{Ar}), 7.22 (d, J = 8.1 Hz, 2H, H_{Ar}), 7.39 (broad, 4H, 2NH_2).

*Compound **3f***

FT-IR (KBr): ν_{max} (cm^{-1}) 3402, 3371, 3129, 2952, 1693, 1663, 1596, 1500, 1377, 1460, 1210. ^{1}H NMR (500 MHz, DMSO-d_6): δ (ppm): 3.02 (s, 6H, 2N–CH_3), 3.35 (s, 3H, N–CH_3), 3.47 (s, 3H, N–CH_3), 3.52 (s, 3H, NMe_2), 3.54 (s, 3H, NMe_2), 5.70 (s, 1H, $CH_{methine}$), 6.78 (d, J = 8.1 Hz, 2H, H_{Ar}), 7.07 (d, J = 8.1 Hz, 2H, H_{Ar}), 7.55 (broad, 2H, NH_2), 7.69 (broad, 2H, NH_2). ^{13}C NMR (125 MHz, DMSO-d_6): δ (ppm): 19.5, 28.5, 30.4, 30.5, 35.0, 40.5, 41.1, 85.4, 113.0, 127.7, 132.1, 148.8, 151.1, 154.3, 164.0. Mass (EI, 70 eV): *m/z* 441 (M^+), 442 (M^++1).

*Compound **3h***

^{1}H NMR (300 MHz, DMSO-d_6): δ (ppm): 3.17 (s, 6H, 2N–CH_3), 3.34 (s, 3H, N–CH_3), 3.36 (s, 3H, N–CH_3), 3.72 (s, 3H, O–CH_3), 5.56 (s, 1H, $CH_{methine}$), 6.77 (d, J = 8.7 Hz, 2H, H_{Ar}), 7.01 (d, J = 8.5 Hz, 2H, H_{Ar}), 7.46 (broad, 4H, 2NH_2); ^{13}C NMR (75 MHz, DMSO-d_6): δ (ppm): 28.4, 30.4, 35.0, 55.4, 86.7, 87.2, 113.6, 128.0, 131.7, 150.9, 154.6, 157.3, 162.3, 163.3.

3. Results and discussion

3.1 Characterization of FSCNSC

The reactions pertaining to the production of [Fe_3O_4 @SiO_2 @Si$(CH_2)_3$–NMe_2–$(CH_2)_2$–NMe_2–SO_3H][Cl]$_2$ (FSCNSC) have been illustrated in Scheme 1 (Experimental section). This magnetic nanocatalyst was characterized by EDS, elemental mapping, FE-SEM, XRD, FT-IR, VSM, TG, and DTG analyses.

As can be seen in Scheme 1, there are carbon, nitrogen, oxygen, iron, silicon, sulfur, and chlorine elements in the structure of FSCNSC; this subject was verified by EDS analysis (Figure 1). In another study, elemental mapping analysis was utilized to show the existing expected elements in the catalyst structure and the dispersion of the elements on its surface (Figure 2). According to this study, all expected elements (carbon, nitrogen, oxygen, iron, silicon, sulfur, and chlorine) are present in the structure of FSCNSC; additionally, this analysis indicates that the mentioned elements are properly dispersed in the catalyst surface.

To determine size and morphology of the catalyst particles, FE-SEM was used; the acquired micrograph is displayed in Figure 3. As it can be seen in the micrograph, the prepared compound is a nanomaterial, since the particles sizes are less than 100 nm (e.g. 13.40, 15.12, 19.51, 25.83 and 64.14 nm). Moreover, the particles have different crystalline forms.

The XRD pattern of FSCNSC was studied in a domain of $2\theta = 5-80°$. The XRD pattern is represented in Figure 4, and the data obtained from the pattern, including peak width (full width at half maximum, FWHM), interplanar distance, the relative intensity of the peaks, and the calculated particle sizes, are illustrated in Table 1. The sharp peaks appeared at 2θ = 30.32, 35.61, 43.33, 53.78, 57.33, and 63.05° corroborated existing a cubic spinel form of Fe_3O_4 in the catalyst structure, and the broad peak at $2\theta \approx 15.70-31.70°$ is ascribed to the SiO_2 layer (amorphous structure). The other peaks are related to diverse crystalline forms of the nanomaterial. The crystallite sizes of FSCNSC, which were calculated by Debye-Scherrer equation $D = K\lambda/(\beta \cos\theta)$, were in the range of $6.76-37.91$ nm and are in good compliance with the FE-SEM results.

The FT-IR spectrums of FSCNSC and the precursors for its synthesis (**I** to **III**) are shown in Figure 5. In the catalyst spectrum, the peaks related to Si–O (rocking) and Fe–O bonds were seen at 469 and 577 cm^{-1}, respectively. The

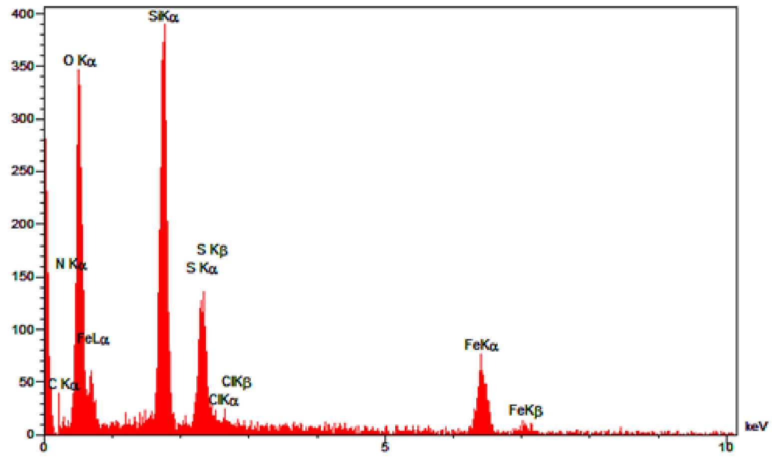


Figure 1. The EDS spectrum of FSCNSC.

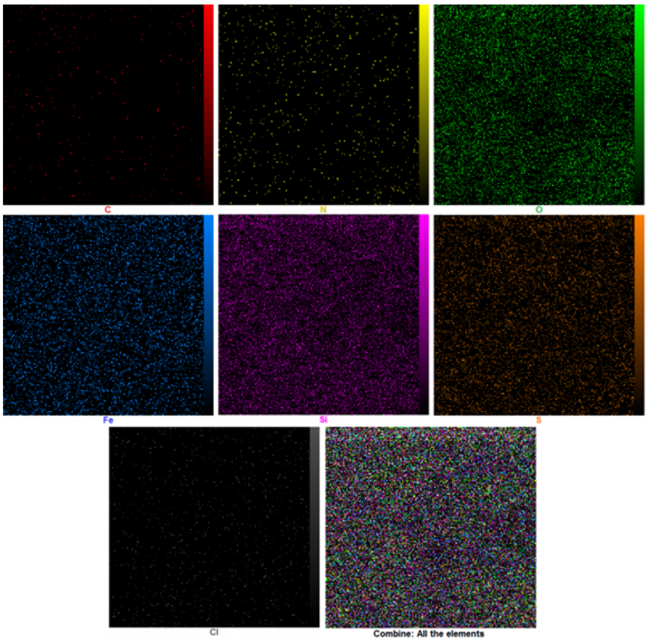


Figure 2. The elemental mapping analysis of FSCNSC.

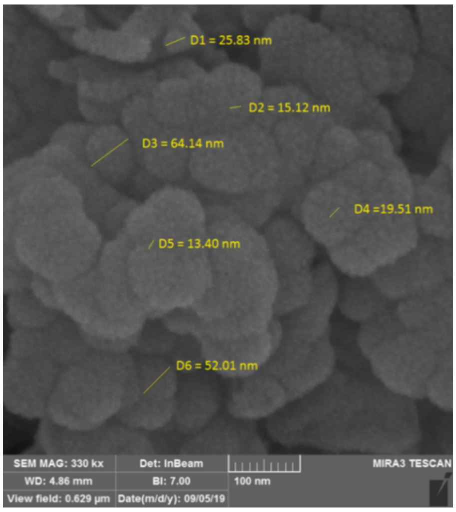


Figure 3. The FE-SEM micrograph of FSCNSC.

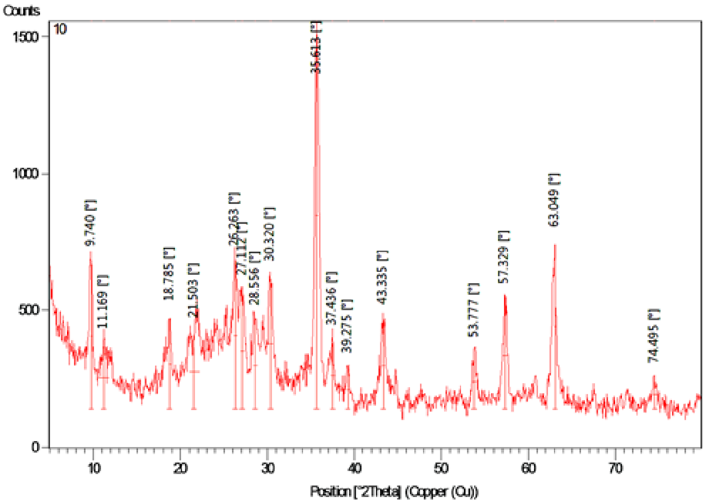


Figure 4. The XRD pattern of FSCNSC.

Table 1. The XRD data of FSCNSC.

2θ (°)	FWHM (°)	Interplanar distance (nm)	Rel. int. (%)	Particle size (nm)
9.7396	0.2952	0.9081	36.67	27.03
11.1695	1.1808	0.7922	15.87	6.76
18.7845	0.2952	0.4724	22.90	27.30
21.5027	1.1808	0.4133	18.77	6.85
26.2634	0.3936	0.3393	36.77	20.74
27.1125	0.3936	0.3289	29.25	20.78
28.5564	0.5904	0.3126	21.82	13.90
30.3204	0.3936	0.2948	33.06	20.93
35.6129	0.2952	0.2521	100.00	28.29
37.4356	0.5904	0.2402	16.80	14.22
39.2747	0.3936	0.2294	9.81	21.45
43.3351	0.3936	0.2088	22.21	21.74
53.7773	0.3936	0.1705	13.63	22.65
57.3295	0.5904	0.1607	27.23	15.35
63.0485	0.2460	0.1474	41.78	37.91
74.4948	0.5904	0.1274	7.26	16.92

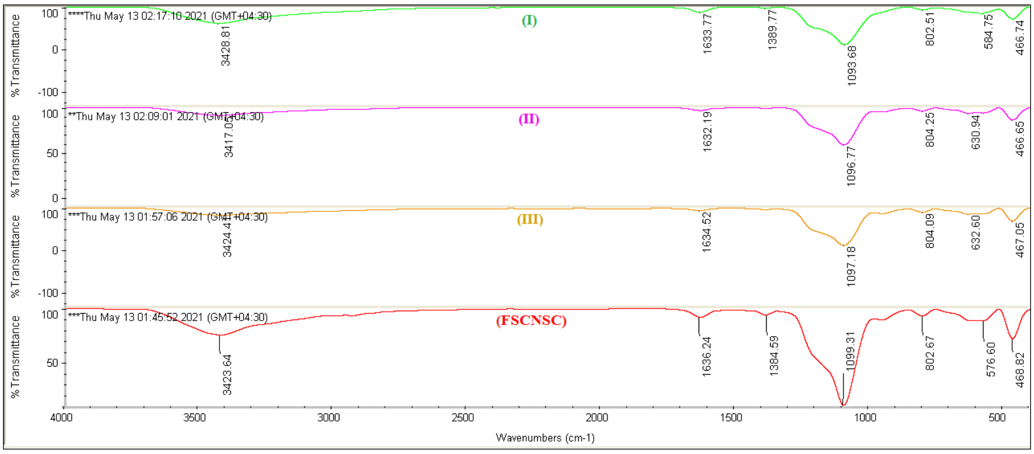


Figure 5. The FT-IR spectrum of the catalyst.

peaks observed at 803 and 1099 cm^{-1} are related to symmetric and asymmetric stretching vibrations of Si–O–Si bonds in the structure of FSCNSC, correspondingly. The peak observed at 1636 cm^{-1} is ascribed to O–H groups in H_2O molecules adsorbed on the SiO_2 surface of the catalyst. The peak relevant to the stretching vibration of the OH group in the SO_3H moiety was observed at $\sim 2680-3685$ cm^{-1}; this peak was not observed in precursors **I** to **III** (this subject confirmed that in the catalyst, the SO_3H group has bonded with NMe_2) (Figure 5). These interpretations of the FT-IR data are in accordance with the literature data [4, 33].

To investigate the magnetic properties of the novel nanomaterial, VSM analysis was utilized (the study was done at room temperature); the attained diagram is exhibited in Figure 6. According to the VSM diagram, saturation magnetization (Ms) of FSCNSC was 24.9 emu/g; Ms of the utilized nano-Fe_3O_4 for its production has been reported 56.7 emu/g [57]. The decrement of Ms of the nanocatalyst in comparison with nano-Fe_3O_4 can be related to coating silica on the nano-Fe_3O_4 and anchoring the organic moieties with the Fe_3O_4@SiO_2 surface. Nevertheless, the catalyst had appropriate magnetization properties and could be magnetically isolated by an external magnet from the reaction mixture. In another investigation, Ms of FSCNSC was compared with compound **III** (the precursor for the catalyst synthesis). Ms of compound **III** was 37 emu/g (we have reported characterization of **III** in our previous article and used it as a basic catalyst) [7]; decreasing Ms of the nanocatalyst in comparison with its previous precursor (**III**) can confirm the successful conversion of **III** to FSCNSC (grafting sulfonic acid moiety to **III**).

To determine the thermal stability of FSCNSC, TGA and DTG analyses were utilized; the relevant diagrams are represented in Figure 7. Weight loss was done in four steps. A slight weight loss was accomplished below ∼150 °C (with Tmax at 111 °C in the DTG diagram); it can be attributed to the vaporization of the adsorbed solvents (and water) on the surface of Fe_3O_4@SiO_2. The second and third weight-losses, which were performed at $\sim 150-380$ (with T_{max} at 300 °C in the DTG diagram) and $380-470$ (with

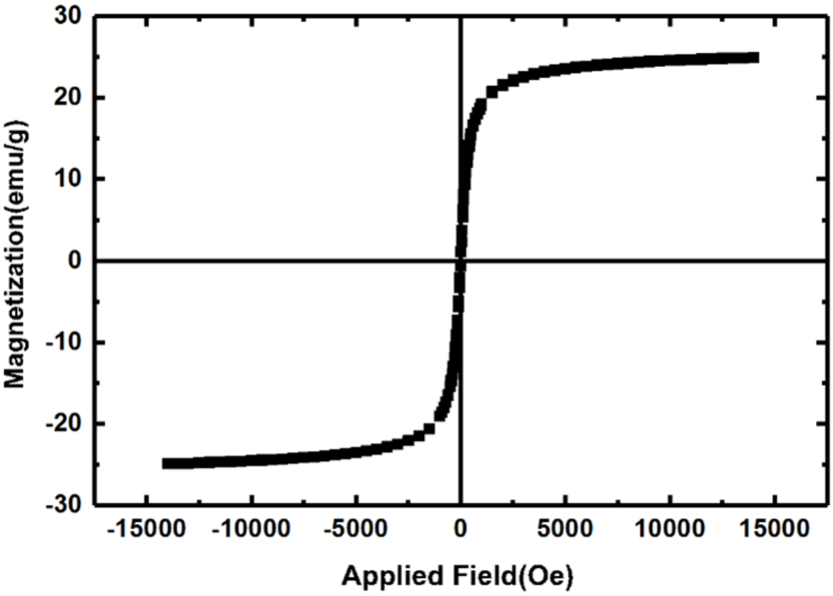


Figure 6. The VSM diagram of FSCNSC.

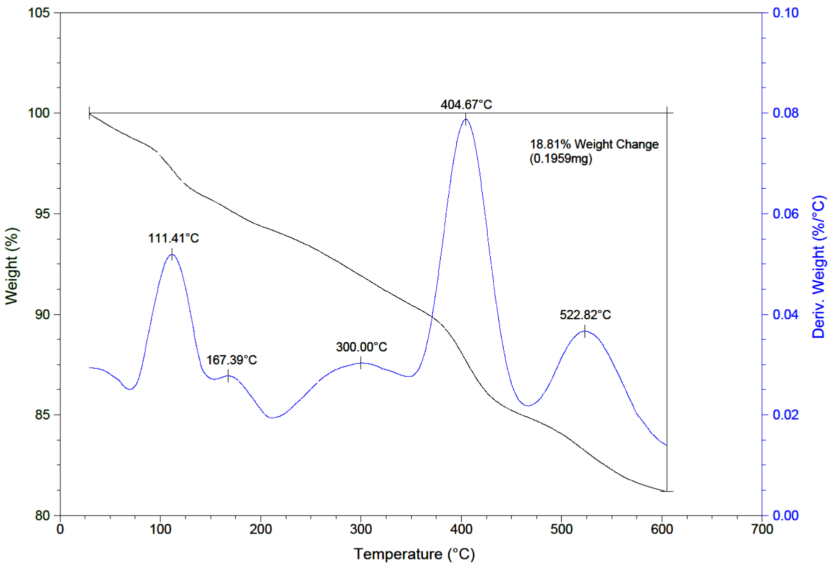


Figure 7. The TG and DTG curves of FSCNSC.

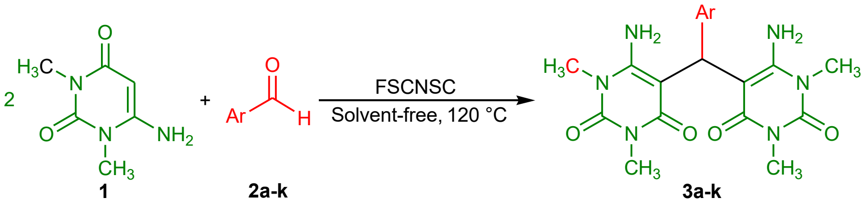
T_{max} at 405 °C in the DTG diagram), can pertain to loss of $CH_2{=}CH_2$, $NHMe_2$, HCl and SO_3 from the organic component grafted with Fe_3O_4@SiO_2, and consequently decomposition of the component. The fourth step of weight loss was done at 470−600 °C (with T_{max} at 523 °C in the DTG diagram) and can be related to the condensation of the silanol groups in SiO_2. The literature data approved these explanations [33, 58, 59]. In another study, the weight-loss percent of FSCNSC (in TG analysis) was compared with that of its precursor (compound **III**). The weight-loss percent of III was 14.13% (we have reported this in our published article [7]), and the weight-loss percent of the catalyst was 18.81%; increasing the weight-loss percent of the catalyst in comparison with compound **III** can approve successful functionalization of **III** by sulfonic acid, and formation of FSCNSC.

3.2 Catalytic performance of FSCNSC for the synthesis of bis(6-amino-1,3-dimethyluracil-5-yl)methanes (3)

In order to determine the most appropriate catalyst amount and the reaction temperature, the condensation of 6-amino-1,3-dimethyluracil (**1**) (2 mmol) with 4-chlorobenzaldehyde (**2c**) (1.05 mmol) was investigated in the presence of different amounts of FSCNSC in the range of 115−125 °C in solvent-free conditions (Scheme 2); the results are briefed in Table 2. The reasonable reaction time and yield were attained when the reaction was performed using 0.06 g of the catalyst at 120 °C (Table 2, entry 2); examining the reaction using fewer amount of the catalyst or at a lower temperature afforded lower yields (Table 2, entries 1 and 4). It is worth noting that enhancing the catalyst amount up to 0.07 g and temperature up to 125 °C slightly decreased the reaction time (Table 2, entries 3 and 5); nevertheless, 0.06 g and

Table 2. Studying the influence of the catalyst amount and temperature on the model reaction.

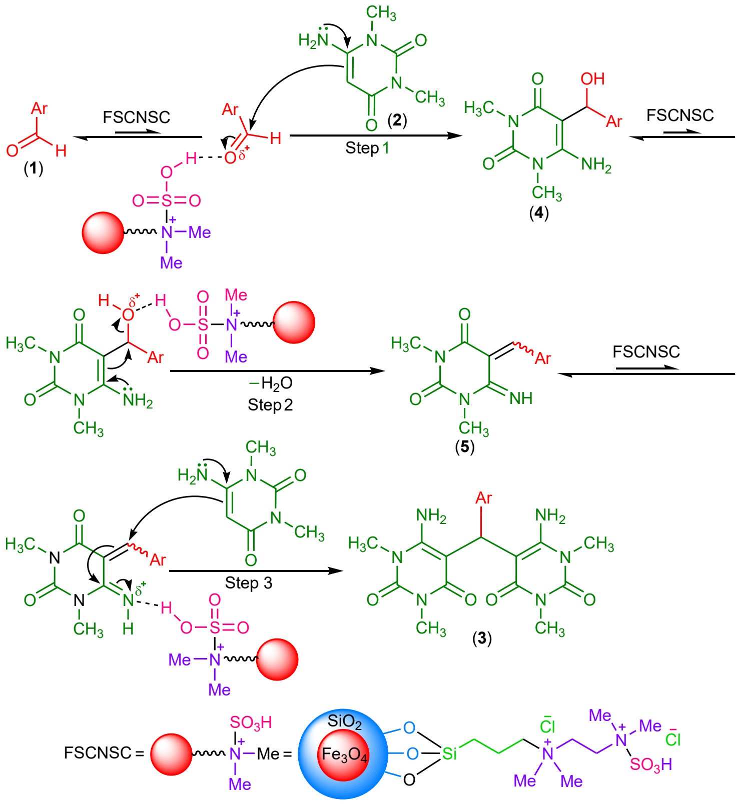
Entry	Catalyst amount (g)	Temp. (°C)	Time (min)	Yield (%)
1	0.05	120	50	92
2	0.06	120	40	96
3	0.07	120	35	96
4	0.06	115	40	93
5	0.06	125	35	96



Scheme 2. The reaction of 6-amino-1,3-dimethyluracil with aryl aldehydes.

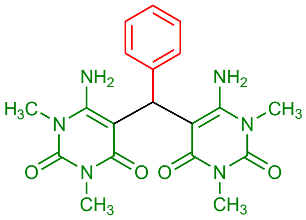
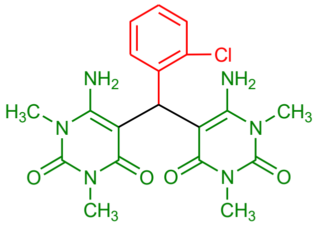
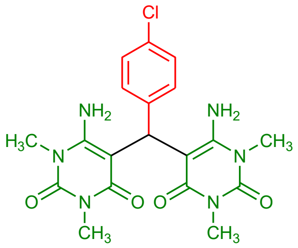
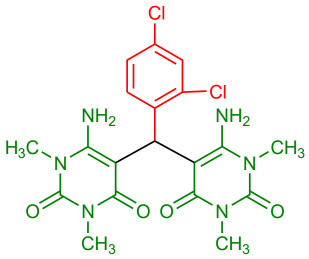
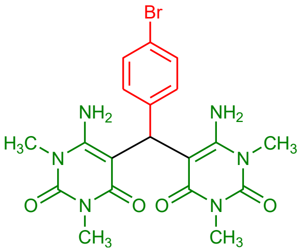
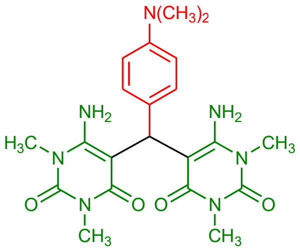
120 °C were chosen as the most appropriate catalyst amount and the reaction temperature, correspondingly, because this is more logical in view of green chemistry principles and economic consideration.

After obtaining the most appropriate reaction conditions, diverse derivatives of bis(6-amino-1,3-dimethyluracil-5-yl)methanes (**3**) were prepared through the reaction of 6-amino-1,3-dimethyluracil (**1**) and various aromatic aldehydes (**2**), in order to assess scope and generality of the procedure. The product structures, reaction times, yields, and melting points of the products (**3**) are demonstrated in Table 3. As can be seen in Table 3, the procedure worked well for aldehydes (**2**) having electron-donating substituents as well as electron-withdrawing ones, and in all cases, high yields of the products (**3**) were attained in relatively short reaction times. So, it can be claimed that FSCNSC is a

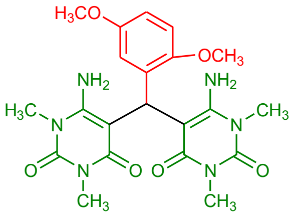
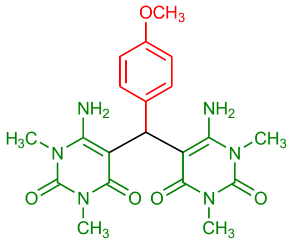
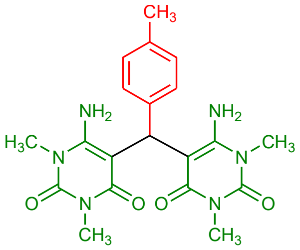
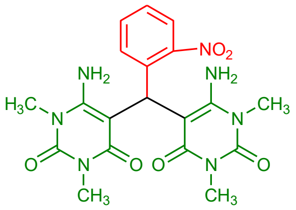
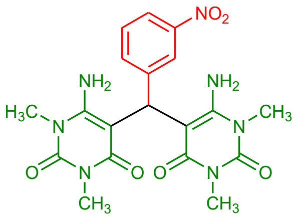


Scheme 3. The proposed mechanism for the FSCNSC catalyzed synthesis of products (**3**).

Table 3. The synthesis of bis(6-amino-1,3-dimethyluracil-5-yl)methane derivatives **(3)** promoted by FSCNSC.

Product No.	Product	Time (min)	Yield (%)[a]	M.p. °C (lit.)
3a		30	96	296-298(294-296) [49]
3b		60	91	257-259(259-260) [47]
3c		40	96	271-273(268-270) [53]
3d		30	88	285-287(283-284) [49]
3e		50	90	262-264(264-266) [47]
3f		60	87	267-269

Continue of Table 3.

Product No.	Product	Time (min)	Yield (%)^a	M.p. °C (lit.)
3g		45	89	268-270(272-274) [53]
3h		30	94	271-273(270-272) [47]
3i		60	96	275-277(272-274) [49]
3j		30	92	263-264(261-263) [47]
3k		60	89	230-232(226-228)[49]

^{a}Isolated yield.

general and effective catalyst for the construction of bis(6-amino-1,3-dimethyluracil-5-yl)methanes **(3)**.

A plausible mechanism was proposed for the construction of bis(6-amino-1,3-dimethyluracil-5-yl)methanes (**3**) (Scheme 3) based on the nucleophilic properties of 6-aminoaminouracils (**1**) [47]. The roles FSCNSC to catalyze the reaction include: (i) accelerating the electrophilic reactions through activation of the electrophiles by its acidic hydrogen (steps 1 and 3), and (ii) facilitating removal of H_2O through conversion of hydroxyl to a good leaving group by the acidic hydrogen (step 2).

Recyclability and reusability of FSCNSC were investigated on the reaction of 6-amino-1,3-dimethyluracil (**1**) with 2-nitrobenzaldehyde (**2j**) to provide compound **3j**; the obtained results are given in Figure 8. The nanocatalyst was recycled according to the procedure given in the experimental section. When the fresh catalyst was used, the reaction time and yield were 30 min and 92%, respectively. In first recycling of FSCNSC, the reaction time and yield were 40 min and 91%, correspondingly. In the second recycling, the reaction time was increased up to 60 min, and the yield was decreased to 83%. Thus, FSCNSC was recyclable and

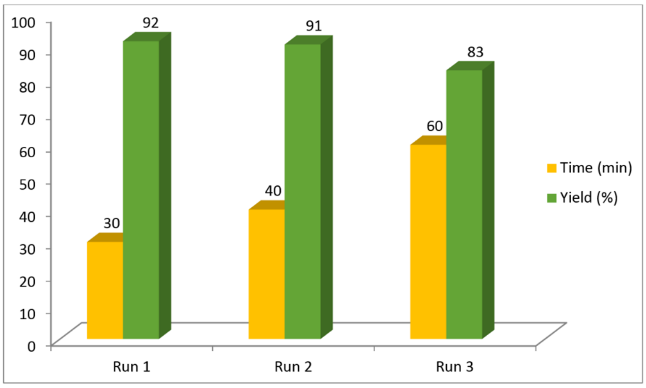


Figure 8. Reusability of FSCNSC on the fabrication of compound **3j**.

reusable for one time with negligible decrement of catalytic activity; however, in the second recycling and reusing, its catalytic activity was partially decreased.

FSCNSC was characterized after recycling using EDS, FT-IR, and FE-SEM analyses. The EDS spectrum of the revised catalyst showed all existing elements in its structure, i.e., C, N, O, Fe, Si, S, and Cl (Figure 9). In the FT-IR spectrum of the recovered catalyst (Figure 10), the peaks belonging to all existing bonds and functional groups in its structure were observed; these peaks consist of 467 (Si–O), 588 (Fe–O), 803 (Si–O–Si), 1095 (Si–O–Si), 1628 (O–H bending of Si–OH), 2926 (aliphatic C–H), and ∼ 3000 – 3727 (O–H group of SO_3H) cm^{-1}. Based on the data attained from EDS and FT-IR analyses, the FSCNSC structure was retained during recycling and reusing. However, the FE-SEM micrograph of the recovered catalyst (Figure 11) illustrated the aggregation of some particles and the formation of larger ones. The decrement in the yield and increment of the reaction time can be related to the aggregation of the catalyst particles and wasting the catalyst during recycling and

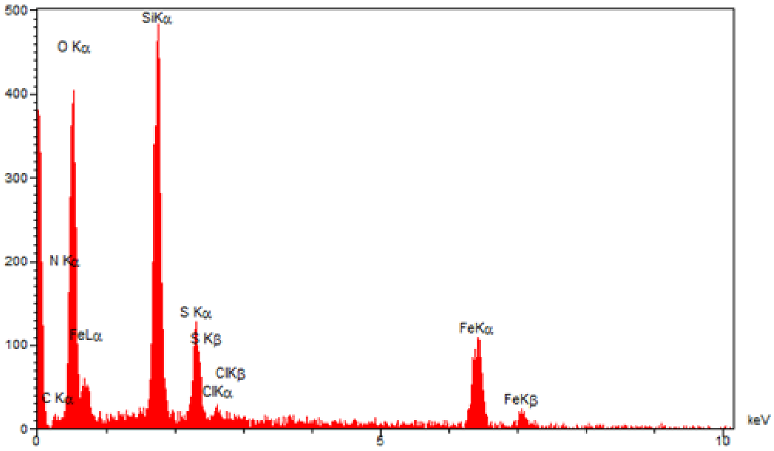


Figure 9. The EDS analysis of the recycled catalyst.

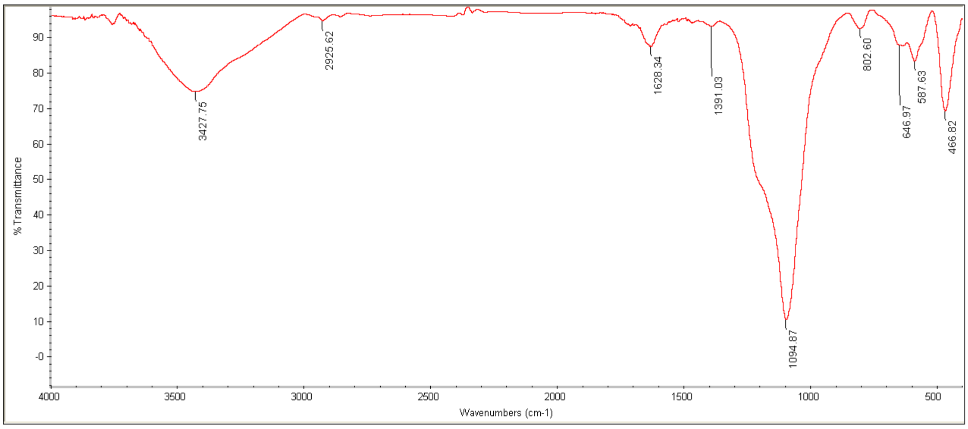


Figure 10. The FT-IR spectrum of the recovered FSCNSC.

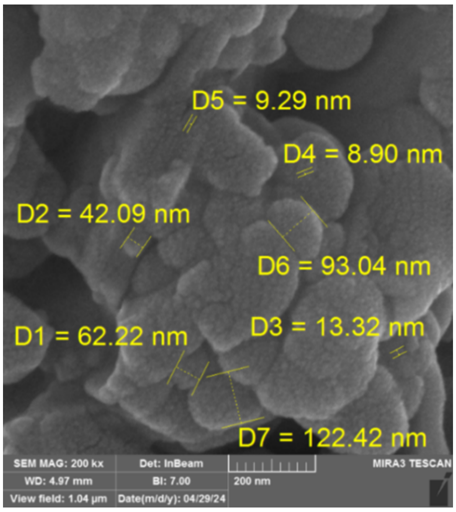


Figure 11. The FE-SEM micrograph of the recycled catalyst.

Table 4. Comparing the reaction conditions and the results of FSCNSC with those of the reported catalysts for the synthesis of products **3a**, **3c**, and **3h**.

Entry	Catalyst	Conditions	Time (min) for products **3a/3c/3h**	Yield (%) for products **3a/3c/3h**
1	FSCNSC	Solvent-free, 120 °C	30/40/30	96/96/94
2	Ceric ammonium nitrate [47]	EtOH/H_2O, r.t.	120/240/360	89/92/89
3	AcOH [48]	MeOH, r.t.	720/–[a]/720	74/–[a]/67
4	$[TMEDA][HSO_4]_2$[b] [49]	Solvent-free, 120 °C	20/20/30	92/95/89
5	SSA-MNPs[c] [50]	EtOH, 50 °C, ultrasound irradiation	60/60/–[a]	87/95/–[a]
6	Catalyst-free [51]	EtOH, reflux	300/270/240	70/72/77
7	Catalyst-free [51]	DMSO, microwave irradiation	3/3/3	90/90/92
8	TEBAC[d] [52]	H_2O, 90 °C	1200-1620[e]	60-98[e]
9	N-[SPIS][TFA][f] [53]	EtOH, reflux	60/60/–[a]	96/95/–[a]

[a] In this research, the compound has not been prepared. [b] *N,N,N′,N′*-Tetramethylethylenediaminium bisulfate. [c] Sulfuric acid functionalized silica-coated magnetic nanoparticles. [d] Triethylbenzylammonium chloride. [e] In this research, compounds **3a**, **3c** and **3h** have not been synthesized; so, the range of the reactions times and yields were presented. [f] Nanorod-[SiO_2-Pr-Im-SO_3H][TFA].

reusing.

In Table 4, the reaction conditions and the efficacy of our nanocatalyst were compared with some reported catalysts; for this comparison, products **3a**, **3c**, and **3h** were chosen. The reaction conditions of our protocol were better than the protocols tabulated in entries 2, 3, 5-7, and 9 (in terms of solvent-free conditions or usage of organic solvents). The reaction times of FSCNSC were shorter than the catalysts displayed in entries 2, 3, 5, 6, 8, and 9. In terms of yields, our catalyst was superior to almost all the catalysts shown in Table 4. Furthermore, we have not used ultrasound and microwave irradiation (which need special apparatuses).

4. Conclusion

In summary, we have prepared a novel magnetic nanomaterial and applied it as a catalyst to fabricate a class of uracil derivatives, namely bis(6-amino-1,3-dimethyluracil-5-yl)methanes. The benefits of this synthesis can be pointed to effectiveness, high yields, relatively short reaction times, usage of few amount of the catalyst, purification of the products by recrystallization, performing the synthesis in solvent-free conditions, recyclability of the catalyst, and adherence to protocols of green chemistry.

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Authors contributions
Authors have equally contributed in preparing the paper.

Availability of data and materials
There is no data available associated with this manuscript.

Conflict of interests
The authors declare that they have no conflict of interest to be declared.

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