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Fabrication of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] as a novel magnetic nanocatalyst, and its utility for the construction of 1,2,4-triazolo[1,5-*a*]pyrimidines

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Original Research

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Abstract:

Nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] (NFSPBF), as a novel magnetic nanomaterial, was fabricated, and its structure was corroborated using EDX, elemental mapping, FE-SEM, XRD, FT-IR, TG, and VSM analyses. In continuation, NFSPBF was employed as an efficacious catalyst for the one-pot multi-component reaction of acetoacetanilide, aryl aldehydes, and 3-amino-1,2,4-triazole to give 1,2,4-triazolo[1,5-*a*]pyrimidines in high yields (86 – 95%) and short times (10 – 15 min). NFSPBF could act as a dual-functional catalyst; $FeCl_4^-$ of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] is a Lewis acid, and its pyridine moiety is a base. It was reusable for two times without a remarkable decrement in its catalytic performance.

Keywords: 3-Amino-1,2,4-triazole; Dual-functional catalyst; Magnetic nanomaterial; Nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$]; 1,2,4-Triazolo[1,5-a]pyrimidine

1. Introduction

Magnetic nanomaterials have several unique characteristics, including environmental friendliness, chemical and thermal sustainability, recoverability, the capacity to manufacture a wide range of them, effortless detachment from the process medium, high surface area, and high performance. Thus, they have been exploited in various industries. Some utilities of magnetic nanomaterials consist of extracting pesticides from fruit juice [1], detaching water/oil [2], electrochemical determining 6-thioguanine (an anticancer agent) [3], preparing supercapacitors [4], degradation of dyes [5], degradating antibiotics [6], electrochemical cell detection [7], manufacturing fuel cells [8], and catalyzing organic reactions [9–22].

Dual-functional catalysts have gained considerable interest because of their high efficiency and adaptability for different applications. These catalysts feature two distinct catalytic sites within their structure, which can work together to boost overall catalytic activity. Various types of such catalysts have been fabricated and used to promote organic transformations [10, 23–27].

"Solvent-free conditions" and "multi-component reactions" are highly effectual, applicatory, economical and ecofriendly approaches that have been broadly applied in organic synthesis, and their profits have been expressed in chemistry sources [28–35].

1,2,4-Triazolo[1,5-a]pyrimidine is an essential constituent of many biological and pharmaceutical substances; for instance, antifungal [36], DNA gyrase inhibitory [37], antibacterial [37], cytotoxic [38], antiproliferative [39], antiepileptic [40], anti-inflammatory [41], anti-HIV-1 [42] and anticonvulsant [43] activities have been reported for these

substances. 1,2,4-Triazolo[1,5-*a*]pyrimidines could be constructed through the one-pot multi-component reaction of β-dicarbonyl compounds, aldehydes, and 3-amino-1,2,4-triazoles; some catalysts have been applied to perform this reaction [44–52].

Having in mind the high significance of magnetic nanocatalysts, 1,2,4-triazolo[1,5-*a*]pyrimidines, solvent-free conditions and multi-component reactions; in this project, we have constructed 1,2,4-triazolo[1,5-*a*]pyrimidines using nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] (NFSPBF) as a novel magnetic nanocatalyst thru a multi-component reaction in the absence of solvent.

2. Experimental

2.1 Materials and apparatuses

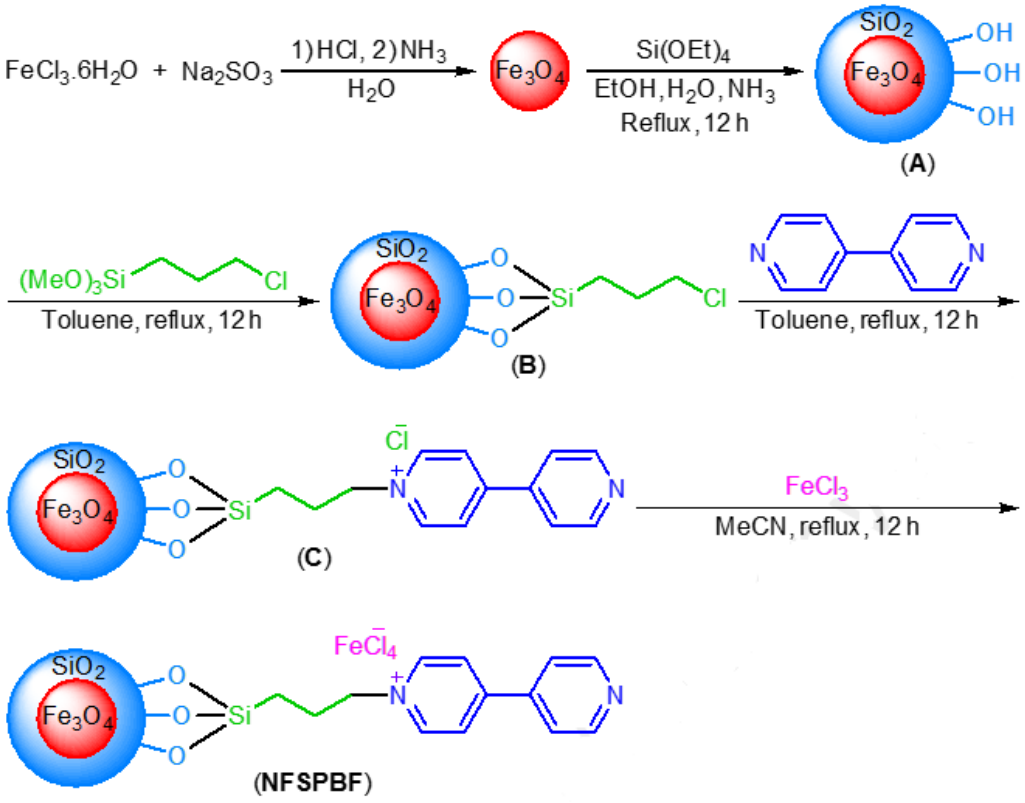
The substances and solvents were provided by Sigma-Aldrich Company. A TESCAN appliance (model MIRA II) was employed for EDX and elemental mapping analyses. FE-SEM pictures were recorded by a TESCAN apparatus (model MIRA III). A Thermo appliance (model AVATAR) was utilized to run the FT-IR spectrum. VSM analysis was accomplished by an MDK (Meghnatis Daghigh Kavir, Iran) at ambient temperature. A TA device (model Q600) was applied to obtain the TG diagram (at $25-600$ °C, with temperature enhancement rate of 10 °C/min in argon atmosphere). XRD analysis was accomplished by a PHILIPS appliance (Cu Kα radiation, $\lambda = 1.54056$ Å, model PW1730). NMR spectra were run on a Bruker Avance DPX FT-NMR apparatus. A Thermo Scientific 9200 appliance was used for the measurement of melting points.

2.2 Fabrication of NFSPBF

Nano-magnetite was fabricated using the procedure reported [53, 54]. Thereafter, nano-magnetite (0.75 g) was added to $Si(OEt)_4$ (2.25 mL) and ammonia (2.40 mL) in H_2O (15 mL) and EtOH (60 mL), and the attained mixture was refluxed and stirred for 12 h to prepare A [54, 55]. (3-Chloropropyl)Trimethoxysilane (0.92 mL, five mmol) was added to A in dry toluene (20 mL), and stirred under reflux conditions for 12 h to synthesize B [54]. In continuation, a mixture of B and 4,4′-bipyridine (0.78 g, 5 mmol) in toluene (20 mL) was stirred under reflux conditions for 12 h to construct C [56]. Lastly, $FeCl_3$ (0.81 g, 5 mmol) was added to C in MeCN (20 mL), and stirred for 24 h under reflux conditions to fabricate NFSPBF. The final step was done according to the literature for similar cases [57, 58]. When the reactants were mixed (for the construction of A, B, C, and NFSPBF), initially, the resulting mixtures were sonicated at room temperature for 20 min, and then refluxed. Moreover, the acquired substance in each stage was magnetically separated, washed with the used solvent in that stage, and dried (Scheme 1).

2.3 The construction of 1,2,4-triazolo[1,5-*a*]pyrimidines

A mixture of acetoacetanilide (0.5 mmol, 0.089 g), aryl aldehyde (0.5 mmol), 3-amino-1,2,4-triazole (0.5 mmol, 0.042 g), and NFSPBF (0.040 g) was strongly stirred by a rod at 90 °C. After consuming the starting substances (as observed by TLC) and cooling the mixture to ambient temperature, EtOAc (15 mL) was added and stirred for 2 min in reflux conditions; NFSPBF was magnetically isolated, washed with EtOAc, and dried. EtOAc of the remainder solution



Scheme 1. The fabrication of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] (NFSPBF).

was distilled, and the residue was recrystallized from EtOH (95%) to fabricate the pure 1,2,4-triazolo[1,5-*a*]pyrimidine.

2.4 Selected NMR data of 1,2,4-triazolo[1,5-*a*]pyrimidines

5-Methyl-7-(4-nitrophenyl)-N-phenyl-4,7-dihydro-[1,2,4]triazolo[1,5-a]pyrimidine-6-carboxamide (e)
^{1}H NMR (300 MHz, DMSO-d$_6$): δ (ppm) 2.22 (s, 3H, CH_3), 6.71 (s, 1H, CH of methine), 7.03 (t, $J = 7.3$ Hz, 1H, H_{Ar}, 7.27 (t, $J = 7.6$ Hz, 2H, H_{Ar}), 7.49 – 7.53 (m, 4H, H_{Ar}), 7.72 (s, 1H, H_{Ar}), 8.21 (d, $J = 8.7$ Hz, 2H, H_{Ar}), 9.83 (s, 1H, NH), 10.45 (s, 1H, NH); ^{13}C NMR (75 MHz, DMSO-d$_6$): δ (ppm) 17.9, 60.1, 103.1, 120.1, 123.92, 124.3, 128.9, 129.1, 137.9, 139.3, 147.7, 148.1, 148.4, 150.8, 165.1.
5-Methyl-7-(4-chlorophenyl)-N-phenyl-4,7-dihydro-[1,2,4]triazolo[1,5-a]pyrimidine-6-carboxamide (g)
^{1}H NMR (300 MHz, DMSO-d$_6$): δ (ppm) 2.20 (s, 3H, CH_3), 6.57 (s, 1H, CH of methine), 7.03 (t, $J = 7.4$ Hz, 1H, H_{Ar}), 7.26 – 7.31 (m, 4H, H_{Ar}), 7.40 (d, $J = 8.4$ Hz, 2H, H_{Ar}), 7.53 (d, $J = 8.1$ Hz, 2H, H_{Ar}), 7.69 (s, 1H, H_{Ar}), 9.79 (s, 1H, NH), 10.33 (s, 1H, NH); ^{13}C NMR (75 MHz, DMSO-d$_6$): δ (ppm) 17.8, 60.1, 103.6, 120.1, 125.9, 128.7, 129.0, 129.1, 129.5, 133.2, 137.3, 140.1, 150.5, 151.3, 165.3.
5-Methyl-7-(2,4-dichlorophenyl)-N-phenyl-4,7-dihydro-[1,2,4]triazolo[1,5-a]pyrimidine-6-carboxamide (h)
^{1}H NMR (500 MHz, DMSO-d$_6$): δ (ppm) 2.17 (s, 3H, CH_3), 6.92 (s, 1H, CH of methine), 7.02 (t, $J = 7.4$ Hz, 1H, H_{Ar}), 7.23 (t, $J = 7.7$ Hz, 2H, H_{Ar}), 7.34 (d, $J = 8.4$ Hz, 1H, H_{Ar}), 7.41 (d, $J = 8.3$ Hz, 1H, H_{Ar}), 7.49 (d, $J = 8.0$ Hz, 2H, H_{Ar}), 7.56 (s, 1H, H_{Ar}), 7.65 (s, 1H, H_{Ar}), 9.85 (s, 1H, NH), 10.35 (s, 1H, NH); ^{13}C NMR (100 MHz, DMSO-d$_6$): δ (ppm) 17.8, 58.1, 102.9, 120.2, 124.0, 128.5, 129.2, 129.7, 132.1, 133.8, 134.2, 137.5, 137.7, 139.5, 148.5, 150.8, 165.1.

3. Results and discussion

3.1 Characterization of NFSPBF

For characterization of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] (NFSPBF), EDX, elemental mapping, FE-SEM, XRD, FT-IR, TG, and VSM analyses were utilized.
The EDX spectrum (figure 1) exhibited the anticipated elemental compositions of NFSPBF, i.e. Fe (related to Fe_3O_4 and $FeCl_4^-$), O (relevant to Fe_3O_4 and SiO_2), Si (pertained to SiO_2 and Si-Pr), C (related to the alkyl chain and the aromatic rings), N (relevant to the aromatic moieties) and Cl (pertinent to $FeCl_4^-$). Moreover, the elemental mapping analysis (figure 2) affirmed the presence of the mentioned elements in the structure of NFSPBF, and good distribution of the elements on the catalyst surface. Based on both analyses, nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] has been successfully fabricated.
FE-SEM was exploited to determine the sizes and morphologies of the particles (figure 3). The gained FE-SEM pictures illustrated that the particles are in nano-size (e.g., 38.37, 41.40, and 42.00 nm), and have quasi-spherical shapes. Moreover, there is porosity between the particles.
In the XRD pattern (figure 4), the peaks appeared at 2θ = 30.41, 35.87, 43.55, 53.98, 57.67, and 63.32º, corroborating the existence of a cubic spinel structure of Fe_3O_4 in the structure of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] [58]. The presence of the amorphous form of the silica shell was confirmed by the observed broad peak at $2\theta \approx 17.00 - 28.80$º [58]. The FWHM (full width at half maximum), interplanar distance, and relative intensity related to each peak, as well as the computed particle sizes of NFSPBF, are indicated in Table 1. The particle sizes (D) were computed by the Debye-Scherrer equation ($D = K\lambda/(\beta\cos\theta)$, where K is the shape factor (0.9), λ is Cu radiation wavelength (0.154178 nm), and β is the FWHM of the peak in radians). The accounted sizes of the particles were in the range of 11.87 – 42.46 nm; these

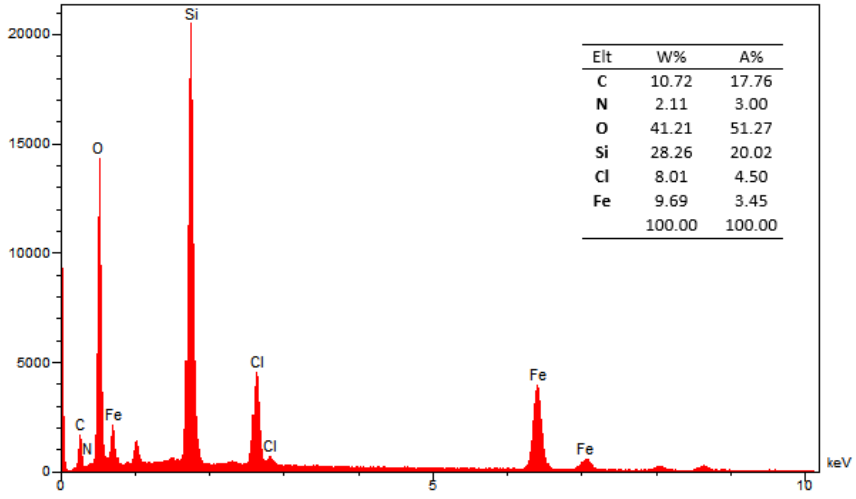


Figure 1. The EDX spectrum of NFSPBF.

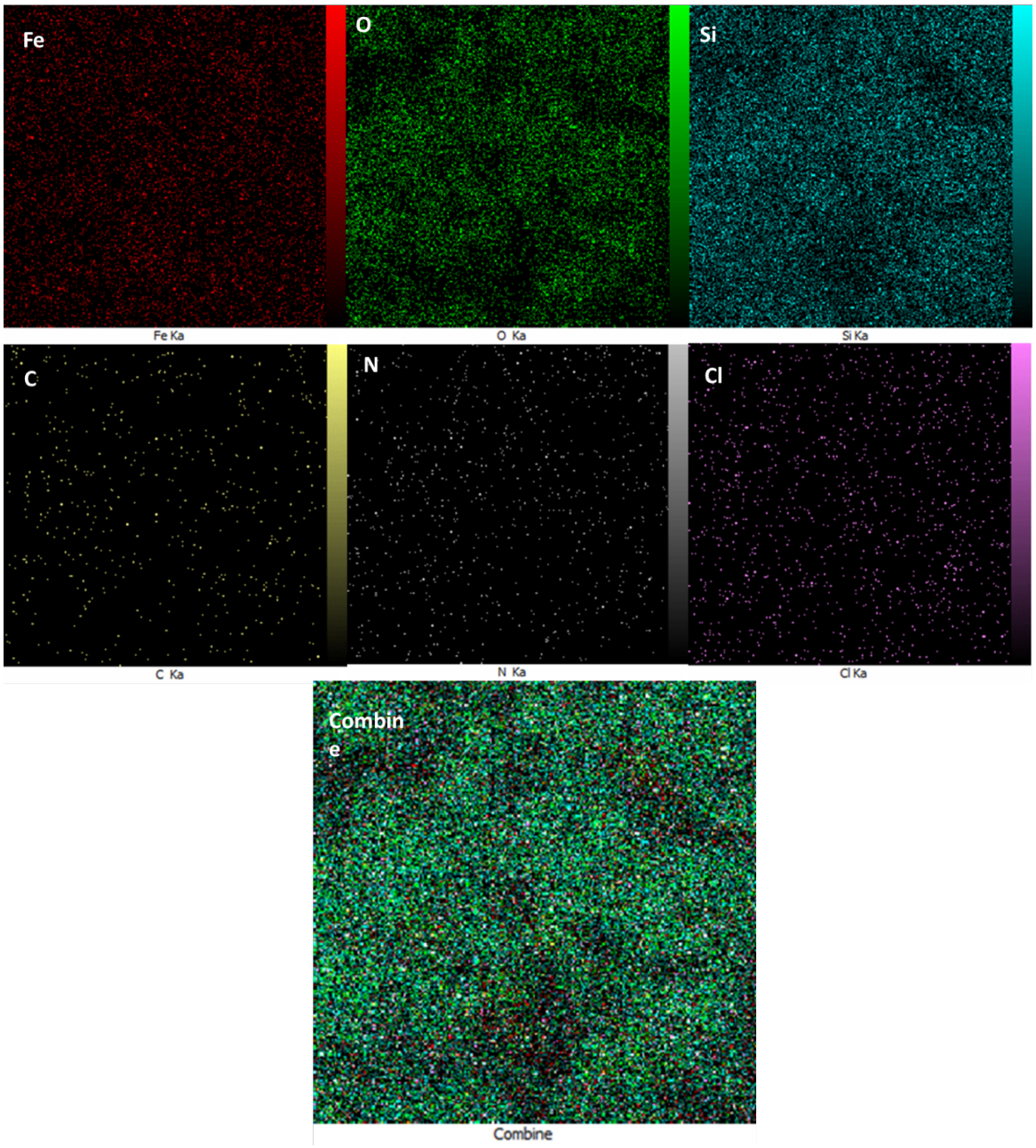


Figure 2. The elemental mapping pictures of NFSPBF.

results were consistent with the ones attained from the FE-SEM pictures.

The FT-IR spectrum of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] is represented in figure 5. The observed band at 471 cm^{-1} is ascribed to the Si–O bond (rocking). The band related to the Fe–O bond was observed at 586 cm^{-1}. The appeared bands at 806 and 1101 cm^{-1} are attributed to symmetric and asymmetric stretching vibrations of Si–O–Si, correspondingly. The O–H bonds of Si–OH groups gave bands at 1635 (bending) and $\sim 3270-3680$ (stretching) cm^{-1}. The band relevant to aliphatic C–H bonds (stretching vibration) was seen at 2925 cm^{-1}.

Thermal constancy of NFSPBF was investigated using TG analysis (figure 6). On the basis of the TG diagram, weight loss of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] has occurred in two stages. First weight loss (up to 200 °C) can be due to vaporization of the absorbed solvents on the nanomaterial surface, and the second one ($200-600$ °C) can be attributed to decomposition of the Si-Pr-Bipyr motif, conversion of $FeCl_4^-$ to $FeCl_3$, oxidation of $FeCl_4^-$, and condensing the silanol groups.

Magnetic behavior of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] and the precursor for its fabrication (compound C in Scheme 1) was investigated by VSM analysis at room temperature; figures 7 and 8 exhibit the corresponding diagrams. Saturation magnetization (Ms) quantities of compound C and NFSPBF were ~ 27.2 and ~ 28.9 emu/g, respectively; the increment in Ms of NFSPBF in comparison with its precursor can be attributed to the successful reaction of C with $FeCl_3$ to produce nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] [59]. Ms of the utilized nano-magnetite for the fabrication of

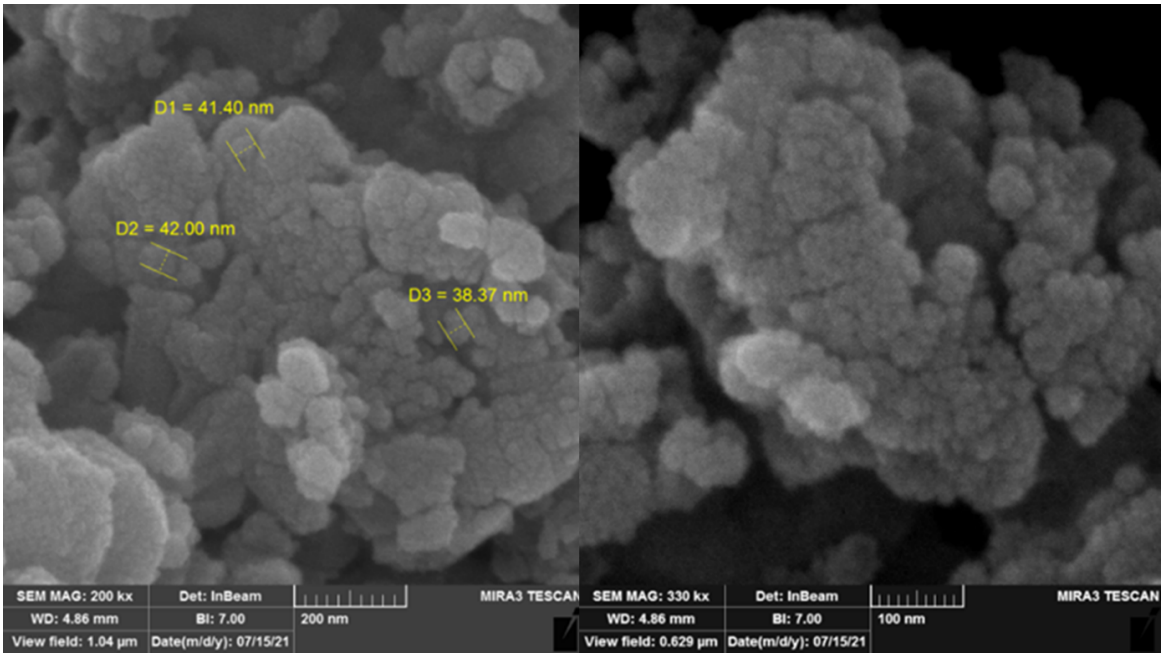


Figure 3. The FE-SEM pictures of the magnetic nanomaterial.

NFSPBF was ∼ 56.7 emu/g [10]; coating silica on the nano-magnetite core, and bonding it to Si-Pr-Bipyr moiety caused a decrease in the Ms of NFSPBF with respect to the nano-magnetite. Nonetheless, NFSPBF had sufficient magnetic properties to be isolated magnetically from the reaction mixture.

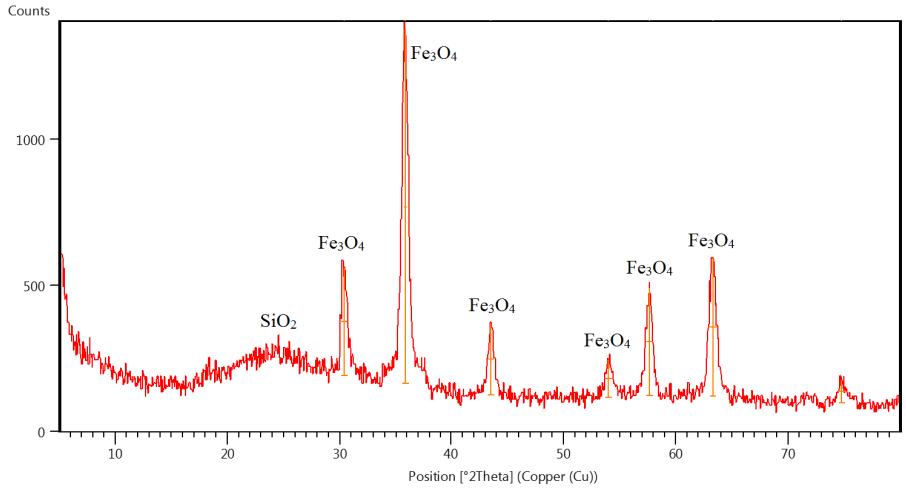


Figure 4. The XRD pattern of NFSPBF.

Table 1. The XRD data of NFSPBF.

2θ (°)	FWHM (°)	Interplanar distance (nm)	Rel. int. (%)	Particle size (nm)
30.4050	0.4920	0.2940	30.54	16.75
35.8693	0.1968	0.2504	100.00	42.46
43.5523	0.3936	0.2078	19.94	21.75
53.9805	0.5904	0.1699	10.65	15.11
57.6747	0.4920	0.1598	30.23	18.45
63.3216	0.7872	0.1469	38.69	11.87
74.7710	0.5904	0.1270	6.27	16.95

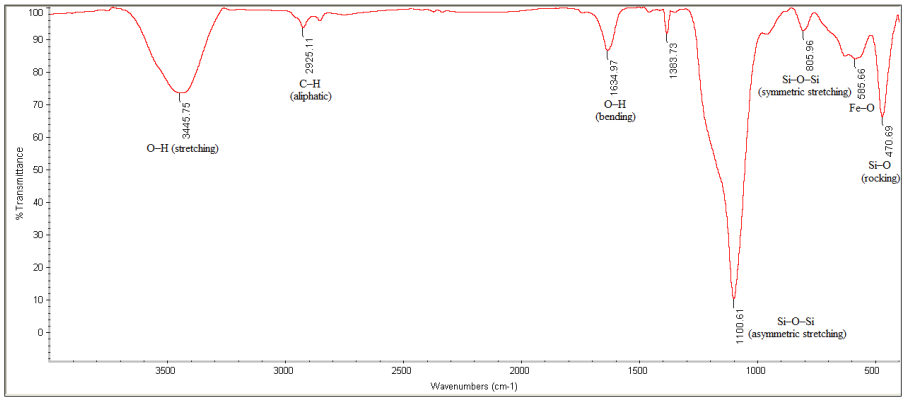


Figure 5. The FT-IR spectrum of the catalyst.

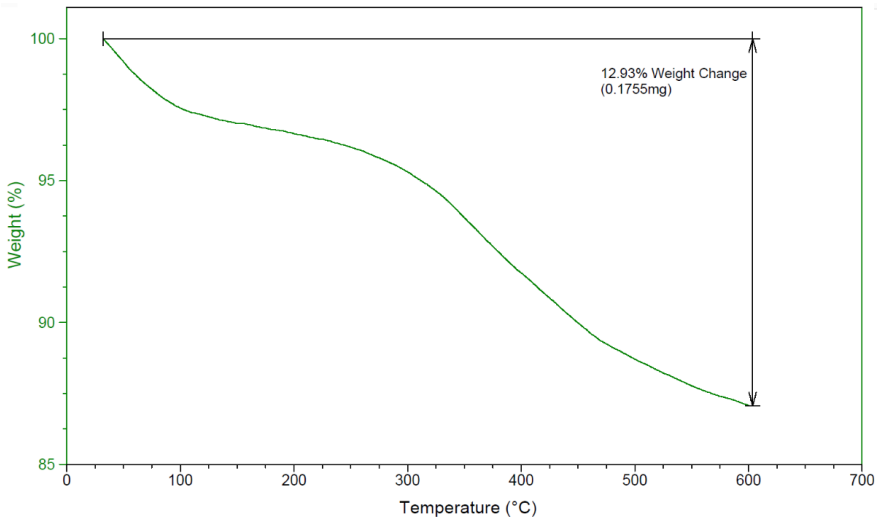


Figure 6. The TG diagram of NFSPBF.

3.2 Utility of NFSPBF for the construction of 1,2,4-triazolo[1,5-*a*]pyrimidines

After full characterization of NFSPBF, we decided to investigate its catalytic capability for the construction of an important category of nitrogen-containing heterocycles, i.e., 1,2,4-triazolo[1,5-*a*]pyrimidines. For this study, the one-pot multi-component condensation of acetoacetanilide (0.5 mmol), 4-nitrobenzaldehyde (0.5 mmol) and 1,2,4-triazole (0.5 mmol) was opted as a model (Scheme 2), and influence of the catalyst amount and temperature on its behavior was studied (the study was done in solvent-free conditions); the results are abridged in Table 2. The relevant 1,2,4-triazolo[1,5-*a*]pyrimidine (e) was constructed in a shorter time with higher yield when the reaction was performed using 0.040 g of NFSPBF at 90 °C (entry 2). The lesser

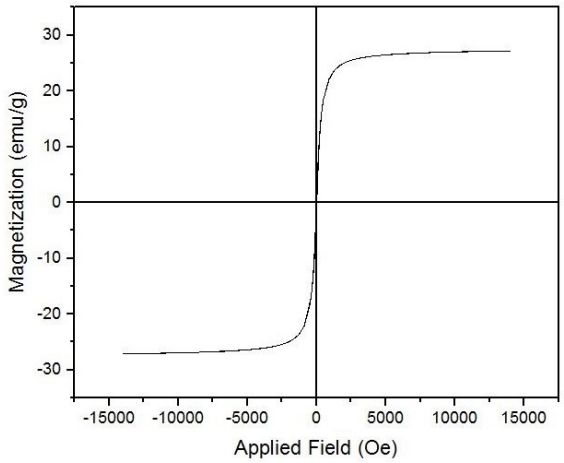


Figure 7. The VSM diagram of compound C.

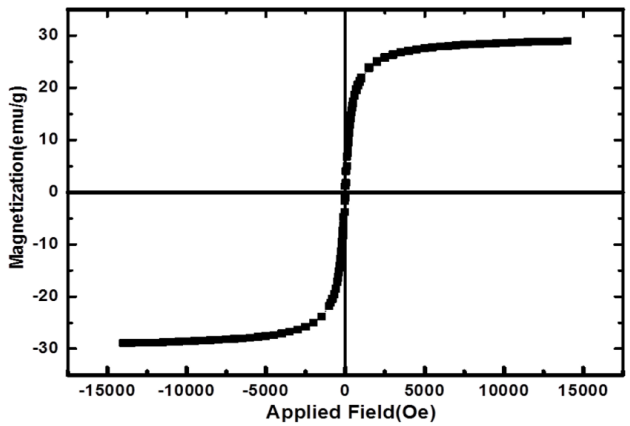
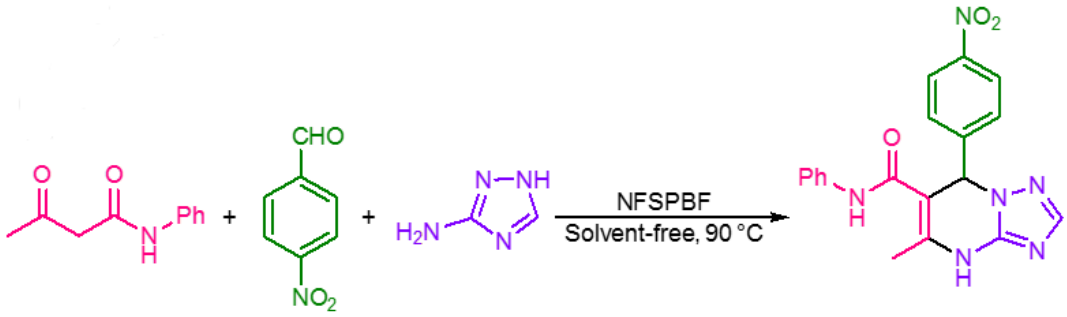


Figure 8. The VSM diagram of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$].



Scheme 2. The model reaction.

catalyst amount and lower temperature could not give satisfactory results (entries 1, 4, and 5). Furthermore, no improvement in the reaction time and yield was observed by enhancement of the catalyst amount up to 0.045 g and the temperature up to 95 °C (entries 3 and 6).

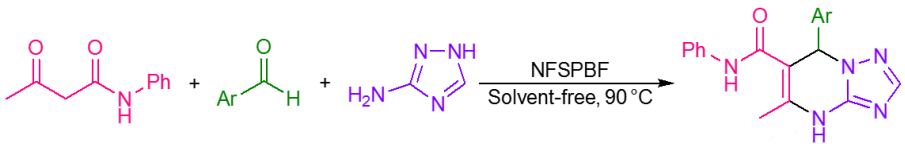
After attaining the optimal conditions, the capability and generality of NFSPBF were appraised for the fabrication of various derivatives of 1,2,4-triazolo[1,5-*a*]pyrimidines by the use of diverse aryl aldehydes in the reaction; Table 3 represents the acquired results. According to the data, aryl aldehydes bearing electron-donating (OH and OCH_3), electron-attracting (NO_2) and halogen (Cl and Br)

Table 2. The results of studying the model reaction.

Entry[a]	Catalyst amount (g)	Temp. (ºC)	Time (min)	Yield (%)
1	0.035	90	15	93
2	0.040	90	10	95
3	0.045	90	10	95
4	0.040	70	25	72
5	0.040	80	20	87
6	0.040	95	10	95

[a]The conditions: acetoacetanilide (0.5 mmol), aldehyde (0.5).

Table 3. The fabrication of 1,2,4-triazolo[1,5-*a*]pyrimidines.



Product No.[a]	Ar	Time (min)	Yield (%)[b]	TON/TOF (h^{-1})	M.p. (ºC) [lit.]
a	C_6H_5	10	92	15.3/91.8	254-256 (253-257) [45]
b	4-HOC_6H_4	15	86	14.3/57.2	275-277 (273-275) [50]
c	4-$CH_3OC_6H_4$	15	94	15.7/62.8	242-244 (245) [47]
d	3-$O_2NC_6H_4$	10	93	15.5/93.0	265-267 (267-269) [46]
e	4-$O_2NC_6H_4$	10	95	15.8/94.8	266-268 (259-261) [45]
f	2-ClC_6H_4	15	89	14.8/59.2	247-249 (250-252) [48]
g	4-ClC_6H_4	10	95	15.8/63.2	265-267
h	2,4-$Cl_2C_6H_3$	15	92	15.3/61.2	248-250 (246) [47]
i	4-BrC_6H_4	10	91	15.2/91.2	228-230 (231-233) [48]

[a]The conditions: acetoacetanilide (0.5 mmol), aldehyde (0.5 mmol), 3-amino-1,2,4-triazole (0.5 mmol), NFSPBF (0.040 g), the reaction medium (solvent-free), and the reaction temperature (90 ºC).

substituents on *para*, *meta* and *ortho* positions afforded the pertaining 1,2,4-triazolo[1,5-*a*]pyrimidines in short times with high yields, and this subject affirmed high capability and generality of NFSPBF to catalyze the reaction.

According to the W% of nitrogen in nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] obtained from the EDX analysis (figure 1), and also considering that only one nitrogen of it is basic and can catalyze the reaction, the amount of basic nitrogen in NFSPBF was found to be ~ 0.75 mmol/g [60] (we have used 6 mol% of the catalyst in the reaction). Based on this amount, turnover number (TON) and turnover frequency (TOF) were calculated for each product (Table 3) TON = yield (%)/mol% of NFSPBF; TOF = TON/the reaction time (h); all 1,2,4-triazolo[1,5-*a*]pyrimidines were constructed with high TON and TOF, and this result also confirmed high efficacy of NFSPBF to catalyze the reaction.

Testing the reusability of NFSPBF was accomplished for the construction of 1,2,4-triazolo[1,5-*a*]pyrimidine e; NFSPBF was recycled using the procedure reported in the experimental section. Figure 9 illustrates the gained results on reusability; no remarkable decrement in the catalytic activity of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] was observed after two times reusing (runs 2 and 3); but, in the third and fourth reuses, its activity was slightly diminished (runs 4 and 5).

NFSPBF was characterized after recycling using FT-IR, FE-SEM, and XRD analyses. In the FT-IR spectrum of the recovered nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] (figure 10), the peaks belonging to all existing bonds and functional groups in its structure were observed; these peaks consist of 468 (Si–O), 589 (Fe–O), 802 (Si–O–Si), 1097 (Si–O–Si), 1633 (O–H bending of Si–OH), ~ 2924 (aliphatic C–H), and ~ 3294 – 3670 (O–H stretching Si–OH) cm^{-1}. Based on the data attained from the FT-IR analysis, the NFSPBF structure was retained during recycling and reusing. The XRD pattern of the recovered NFSPBF (figure 11) illustrated sharp peaks at $2\theta \approx 30.44$, 35.83, 43.54, 53.94, 57.44 and 63.12°, and a broad peak at $2\theta \approx 19.10 - 28.90$°; the sharp peaks is ascribed to Fe_3O_4 in the NFSPBF structure, and the broad peak is pertained to the amorphous structure of the silica shell. However, considering the FE-SEM micrographs of the recovered NFSPBF (figure 12), it seems that some particles were aggregated and formed larger ones. The decrement of the yield and increment of the reaction time can be related to aggregation of the catalyst particles and wasting the catalyst during recycling and reusing.

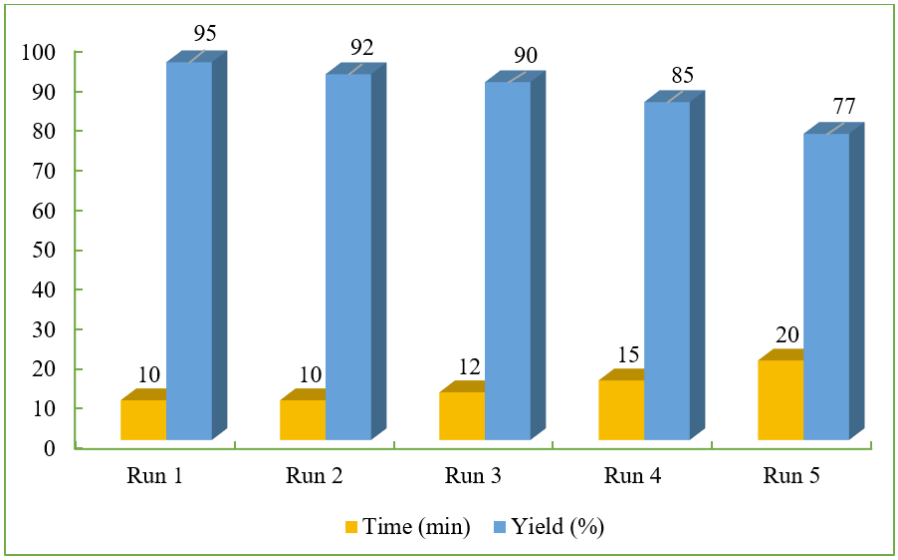


Figure 9. The reusability results of NFSPBF.

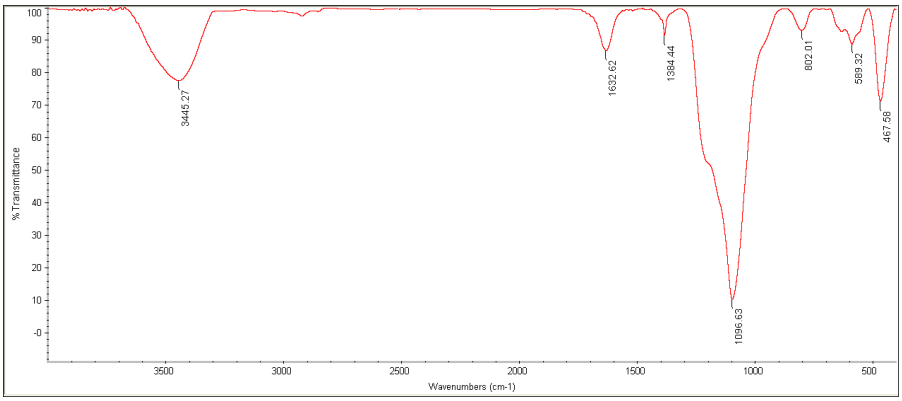


Figure 10. The FT-IR spectrum of the recovered NFSPBF.

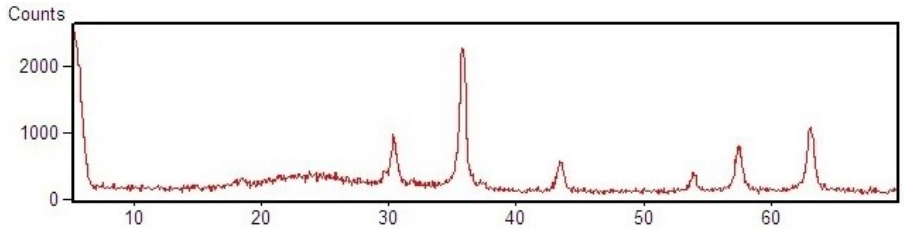


Figure 11. The XRD pattern of the recovered NFSPBF.

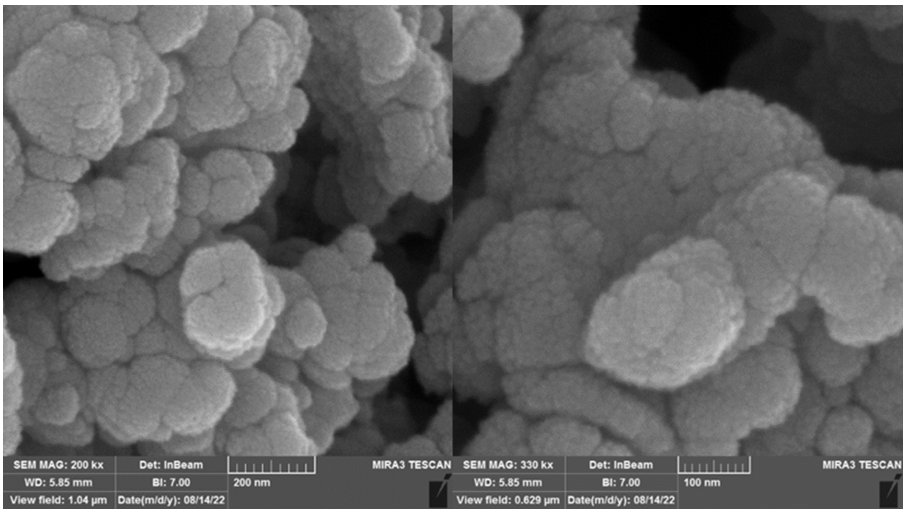


Figure 12. The FE-SEM micrographs of the recovered NFSPBF.

$FeCl_4^-$ of nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$] is a Lewis acid [61, 62], and its pyridine moiety is a base, so, it is a dual-functional (acidic-basic) catalyst; based on this matter and the literature [44, 48], a reasonable mechanism was designed for the construction of 1,2,4-triazolo[1,5-*a*]pyrimidines using NFSPBF (Scheme 3). The catalyst duties are clarified in the mechanism; the electrophiles can be activated by $FeCl_4^-$ to accept nucleophiles (steps 1, 3, and 4), and the nucleophiles can be activated through proton abstraction by the pyridine moiety (steps 1, 3, and 4). Moreover, H_2O removal can be performed by both acidic and basic sites of NFSPBF (steps 2 and 5).

4. Conclusions

In brief, we have introduced a novel dual-functional magnetic nanocatalyst, namely nano-[Fe_3O_4@SiO_2@Si-Pr-Bipyr][$FeCl_4$]; it may catalyze organic reactions by its basic (pyridine moiety) and Lewis acidic ($FeCl_4^-$) sites. We have accomplished the fabrication of 1,2,4-triazolo[1,5-*a*]pyrimidine derivatives from acetoacetanilide, aryl aldehydes and 3-amino-1,2,4-triazole using our nanocatalyst; the protocol has several privileges, consisting of high capability, generality, short reaction times, usage of solvent-free technique and a multi-component reaction, high yields, simplicity (performing the reaction, workup and purifying the products), magnetic recoverability of the catalyst and good agreement with principles of green chemistry.

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Authors contributions

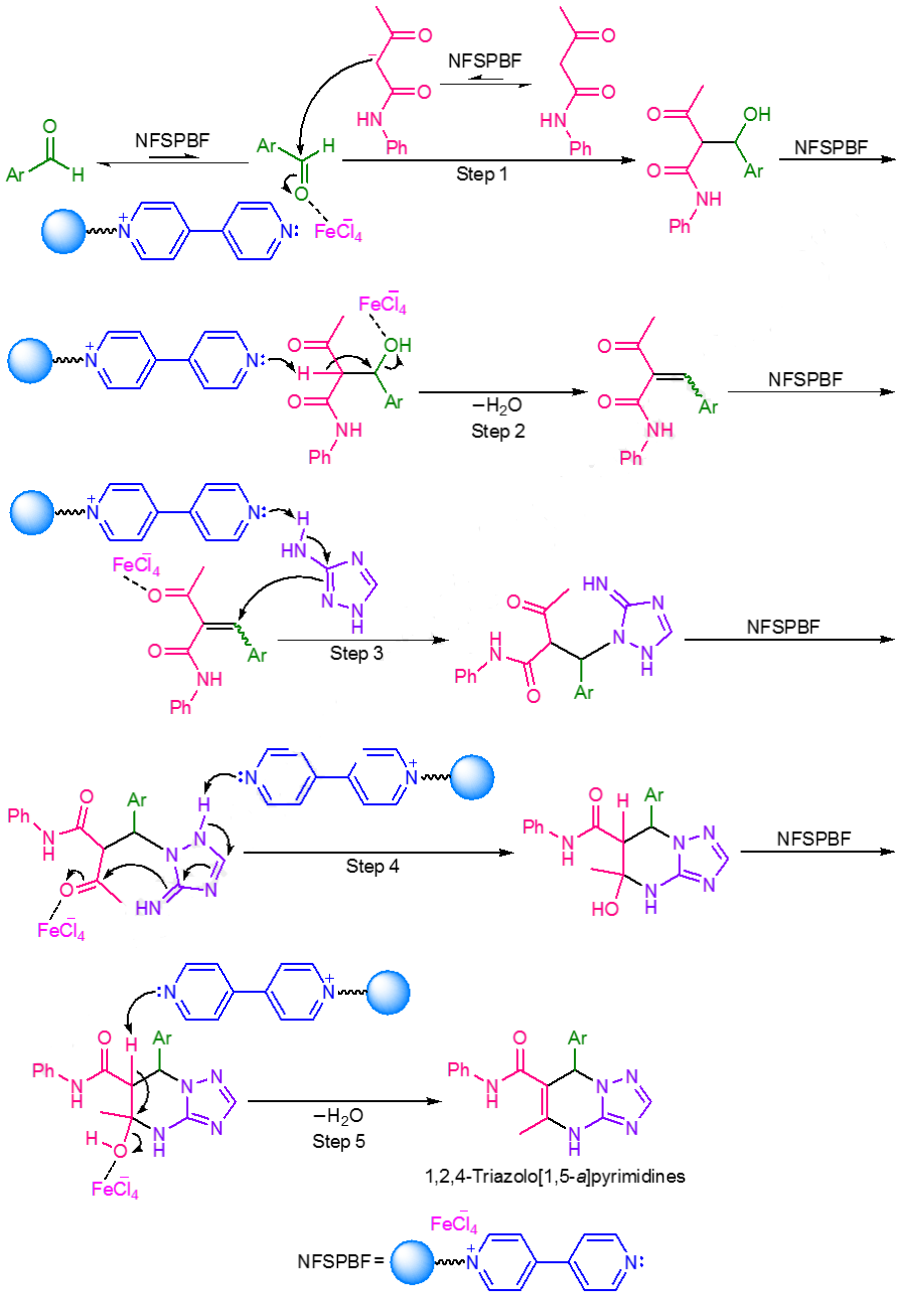
All authors contributed to the study conception. Aboolkarim Zare designed the project. Data collection (through experiments) was done by Elham Jazinizadeh. Interpretation of the analyses were performed by all authors. The first draft of the manuscript was written by Aboolkarim Zare, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Availability of data and materials

The data that support the findings of this study are available from the corresponding author, upon reasonable request.

Conflict of interests

The author declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.



Scheme 3. The reaction mechanism.

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