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Fe_3O_4@GO-Pr-SO_3H AS AN EFFICIENT AND RECYCLABLE CATALYST FOR THE SYNTHESIS OF PYRIDINE DICARBONITRILES

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ABSTRACT
Fe_3O_4@GO-Pr-SO_3H (FGOSA) has applied as nano-catalyst for fast and clean synthesis of pyridine dicarbonitriles by one-pot multicomponent reaction of 4-methyl thiophenol, malononitrile, and aryl aldehydes. The final outcomes have shown that FGOSA exhibited great catalytic performance for the preparation of pyridine dicarbonitrile products. Furthermore, the catalyst was recyclable and could be reused at least three times without any discernible loss in its catalytic activity. Besides, this green procedure provides prompt achievement to the appropriate products with acceptable yields in ethanol as solvent at reflux situation with easy work-up process.

KEYWORDS: Pyridine dicarbonitriles; Multicomponent reaction; Fast synthesis; Fe_3O_4@GO-Pr-SO_3H

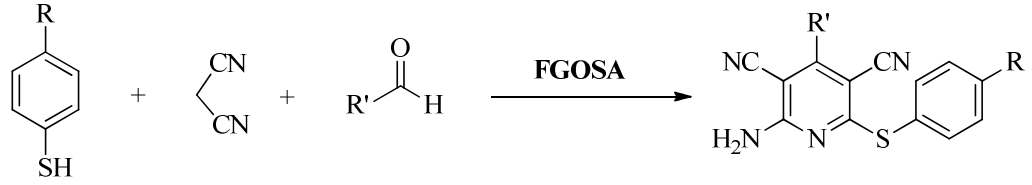
INTRODUCTION
Among the nitrogen-containing heterocycles, densely substituted pyridine derivatives are one of the most important classes of compounds as they widely occur as key structural subunits in numerous natural products that exhibit many interesting biological activities[i]. In addition, these heterocyclic compounds have found a variety of applications in medicinal and pharmaceutical sciences[ii]. Among these pyridine derivatives, 2-amino-6-(arylthio)pyridine-3,5-dicarbonitrile is a privileged scaffold for developing pharmaceutical agents because various compounds with this structural motif display significant and diverse biological activities. For examples, adenosine receptors are associated with Parkinson's disease, hypoxia, asthma, epilepsy, cancer, and cardiovascular diseases[iii]. These pyridine compounds have been shown to be active inhibitors of the adenosine receptors and, therefore, can be used for treating these diseases[iii]. They are also

inhibitors of cholinesterases and may be used for treating neurodegenerative diseases. These compounds have also been studied as potential anti-HBV, anti-bacterial, antibiofilm, and anti-infective agents and as potassium channel openers with applications in treating urinary incontinence. Moreover, some of these derivatives also inhibit prion replication and may be used for treating Creutzfeldt–Jacob disease[iv].

Many synthetic protocols were developed to accelerate the synthesis rate of pyridine dicarbonitriles reaction and to improve the yield. These compounds have been synthesized in the presence of various catalysts such as Et_3N[v], IBX[vi], DABCO[vii], K_2CO_3[viii], KOH[ix], MS 4A[x], Nano-CuI[xi], Nano-MgO[xii], have been utilized in the preparation of the pyridine dicarbonitriles compounds. Major drawbacks of these procedures include expensive reagents, use of large amounts of organic solvents, prolonged heating and side reactions.

In recent years, economic and environmental concerns encourage the application of heterogeneous catalysts to carry out various organic transformations[xiii]. These catalysts have a very high catalytic activities due to their large specific surface area and make the processes clean, safe, high-yielding and inexpensive[xiv]. Nowadays, application of magnetic nanocomposites as heterogeneous catalysts is an interesting research area. The surface functionalization of these materials is an elegant way to bridge the gap between heterogeneous and homogeneous catalyses[xv].

As a part of our research in developing efficient methods of organic synthesis that involve reusable catalysts[xvi-xxxi], we studied application of covalently bonded sulfonic acid magnetic graphene oxide (Fe_3O_4@GO-Pr-SO_3H) as an efficient acid catalyst in the synthesis of pyridine dicarbonitrile derivatives by the reaction of thiophenols **1**, malononitrile **2**, and aryl aldehydes **3**. Nano- Fe_3O_4@GO-Pr-SO_3H (FGOSA) could be readily separated from the reaction mixture by a permanent magnet and reused several times. The process is more effective than filtration and centrifugation in preventing loss of the solid catalyst. However, there were no reports on application of FGOSA as an acidic heterogeneous catalyst in the synthesis of pyridine dicarbonitriles (Scheme 1).



1 **2** **3** **4**

Scheme 1. Synthesis of pyridine dicarbonitriles catalyzed by FGOSA.

EXPERIMENTAL

All chemicals were purchased from Fluka (Buchs, Switzerland) or Merck Companies and used without further purification. FT–IR spectra were recorded for KBr disks on a Tensor 27 Bruker Spectrophotometer. ^1H and ^{13}C NMR spectra were measured on a BRUKER DRX-500 AVANCE spectrometer in DMSO or $CDCl_3$ as solvent. GC/MS spectra were recorded on an Agilent Technologies 6890 network GC system and an Agilent 5973 network Mass selective detector.

General experimental procedure

To a magnetically stirred solution of aldehyde (1 mmol), 4-methyl thiophenol (1 mmol), malononitrile (2 mmol), and FGOSA (0.06 g), 5 mml of ethanol was added and reflux for appropriate time. The reaction was monitored by TLC. Upon completion of the transformation

the catalyst was filtered under hot conditions. The catalyst was separated using an external magnet and washed with hot ethanol (10 mL). After cooling, the combined filtrate was allowed to stand at room temperature. The precipitated solid was collected by filtration, and recrystallized from ethanol to give desired compound in high yields.

2-amino-4-phenyl-6-(p-tolylthio)pyridine-3,5-dicarbonitrile (4a): IR (KBr) (ν_{max}/cm^{-1}): 3432, 3374, 3073, 2922, 2231, 1621, 1585, 1456, 1073, 723; ^{1}H NMR (DMSO, 500 MHz) (δ, ppm): 2.38 (s, 3H, CH_3), 7.32 (d, *J* = 7.1 Hz, 2H, aromatic H), 7.33-7.55 (m, 5H, aromatic H), 7.57 (d, *J* = 7.3 Hz, 2H, aromatic H), 7.60 (s, 1H, aromatic H), 7.76 (br., s, 2H, NH_2); ^{13}C NMR (DMSO, 125 MHz) (δ, ppm): 167.51, 161.12, 159.31, 141.23, 135.54, 134.63, 135.21, 133.89, 130.11, 129.54, 128.89, 124.37, 116.58, 115.87, 95.21, 86.25, 22.48; GC/MS: 341 (M^+).

2-amino-4-(4-chlorophenyl)-6-(p-tolylthio)pyridine-3,5-dicarbonitrile (4b): IR (KBr) (ν_{max}/cm^{-1}): 3421, 3374, 3101, 2965, 2219, 1647, 1585, 1421, 1225, 852 ^{1}H NMR ($CDCl_3$, 500 MHz) (δ, ppm): 2.39 (s, 3H, CH_3), 7.28 (d, *J* = 6.9 Hz, 2H, aromatic H), 7.30-7.40 (m, 2H, aromatic H), 7.43-7.60 (m, 5H, aromatic H), 7.79 (br., s, 2H, NH_2); ^{13}C NMR ($CDCl_3$, 125 MHz) (δ, ppm): 170.21, 160.23, 159.21, 142.32, 137.23, 136.54, 132.65, 130.30, 130.01, 129.11, 122.99, 116.32, 115.54, 97.01, 87.63, 23.29; GC/MS: 375 (M^+).

2-amino-4-(4-nitrophenyl)-6-(p-tolylthio)pyridine-3,5-dicarbonitrile (4c): IR (KBr) (ν_{max}/cm^{-1}): 3503, 3458, 3058, 2968, 2214, 1652, 1565, 1497, 1341, 1254, 1037, 789; ^{1}H NMR (DMSO, 500 MHz) (δ, ppm): 2.26 (s, 3H, CH_3), 7.36 (d, *J* = 7.1 Hz, 2H, aromatic H), 7.39 (d, *J* = 7.1 Hz, 2H, aromatic H), 7.40-7.75 (m, 5H, aromatic H), 8.31 (br, s, 2H); ^{13}C NMR (DMSO, 125 MHz) (δ, ppm): 165.73, 160.71, 158.21, 137.72, 136.01, 134.92, 132.21, 130.42, 129.33, 128.52, 126.21, 116.31, 114.71, 91.95, 88.27, 24.10; GC/MS: 386 (M^+).

2-amino-4-(3-bromophenyl)-6-(p-tolylthio)pyridine-3,5-dicarbonitrile (4d): IR (KBr) (ν_{max}/cm^{-1}): 3441, 3326, 3025, 2998, 2231, 1678, 1589, 1521, 1229, 998, 796; ^{1}H NMR (DMSO, 500 MHz) (δ, ppm): 2.37 (s, 3H, CH_3), 7.31 (d, *J* = 6.9 Hz, 2H, aromatic H), 7.35-7.75 (m, 7H, aromatic H), 7.81 (br, s, 2H, NH_2); ^{13}C NMR (DMSO, 125 MHz) (δ, ppm): 167.56, 162.25, 158.58, 142.20, 140.99, 138.55, 135.89, 134.11, 133.22, 131.78, 130.41, 125.25, 124.31, 116.28, 115.66, 94.52, 87.66, 20.73; GC/MS: 420 (M^+).

2-amino-4-(3-chlorophenyl)-6-(p-tolylthio)pyridine-3,5-dicarbonitrile (4e): IR (KBr) (ν_{max}/cm^{-1}): 3456, 3372, 3099, 2903, 2221, 1671, 1559, 1486, 1001, 803; ^{1}H NMR ($CDCl_3$, 500 MHz) (δ, ppm): 2.47 (s, 3H, CH_3), 7.32 (t, *J* = 7.1 Hz, 2H, aromatic H), 7.38 (d, *J* = 7.3 Hz, 2H, aromatic H), 7.40-7.55 (m, 4H, aromatic H), 7.57 (s, 1H, aromatic H), 7.80 (br., s, 2H, NH_2); ^{13}C NMR ($CDCl_3$, 125 MHz) (δ, ppm): 169.89, 159.21, 157.92, 143.13, 142.03, 139.28, 136.67, 135.09, 132.54, 130.98, 130.85, 128.62, 125.81, 116.52, 115.11, 96.11, 88.23, 22.13; GC/MS: 375 (M^+).

2-amino-4-(3-nitrophenyl)-6-(p-tolylthio)pyridine-3,5-dicarbonitrile (4f): IR (KBr) (ν_{max}/cm^{-1}): 3413, 3258, 3023, 2969, 2245, 1639, 1528, 1468, 1247, 985, 729; ^{1}H NMR (DMSO, 500 MHz) (δ, ppm): 2.79 (s, 3H, CH_3), 7.33 (d, *J* = 7.3 Hz, 2H, aromatic H), 7.47 (d, J = 7.3 Hz, 2H, aromatic H), 7.61 (s, 1H, aromatic H), 7.66 (m, 1H, aromatic H), 7.68 (d, *J* = 7.1 Hz, 1H, aromatic H), 7.69-7.78 (m, 2H, aromatic H), 8.13 (br., s, 2H, NH_2); ^{13}C NMR (DMSO, 125 MHz) (δ, ppm): 166.33, 159.52, 158.12, 141.23, 137.48, 136.20, 135.41, 133.77, 132.71, 131.01, 129.66, 128.21, 126.27, 116.89, 114.22, 95.21, 88.01, 23.11; GC/MS: 386 (M^+).

2-amino-4-(4-methoxyphenyl)-6-(p-tolylthio)pyridine-3,5-dicarbonitrile (4g): IR (KBr) (ν_{max}/cm^{-1}): 3468, 3329, 3029, 2909, 2215, 1667, 1559, 1545, 1459, 1371, 1073, 868; ^{1}H NMR ($CDCl_3$, 500 MHz) (δ, ppm): 2.37 (s, 3H, CH_3), 3.91 (s, 3H, OCH_3), 7.10-7.20 (m, 4H,

aromatic H), 7.34 (d, *J* = 7.2 Hz, 2H, aromatic H), 7.56 (s, 1H, aromatic H), 7.66 (d, *J* = 7.2 Hz, 2H, aromatic H), 7.72 (br., s, 2H, NH_2); ^{13}C NMR ($CDCl_3$, 125 MHz) (δ, ppm): 167.51, 161.42, 159.72, 142.21, 136.61, 133.20, 132.06, 131.13, 127.33, 125.03, 122.62, 115.33, 113.42, 93.66, 88.63, 22.33; GC/MS: 371 (M^+).

2-amino-4-(4-bromophenyl)-6-(p-tolylthio)pyridine-3,5-dicarbonitrile (4h): IR (KBr) (ν_{max}/cm^{-1}): 3441, 3336, 3122, 3962, 2231, 1628, 1571, 1560, 1006, 811; ^1H NMR (DMSO, 500 MHz) (δ, ppm): 2.35 (s, 3H, CH_3), 7.30-7.70 (m, 6H, aromatic H), 7.75 (s, 1H, aromatic H), 7.79 (d, *J* = 7.1 Hz, 2H, aromatic H), 7.83 (br., s, 2H, NH_2); ^{13}C NMR (DMSO, 125 MHz) (δ, ppm): 167.65, 161.58, 158.41, 141.52, 135.82, 134.14, 133.03, 132.69, 131.26, 125.45, 124.65, 117.01, 115.21, 93.96, 88.11, 22.07; GC/MS: 420 (M^+).

2-amino-4-(4-chlorophenyl)-6-(phenylthio)pyridine-3,5-dicarbonitrile (4i): IR (KBr) (ν_{max}/cm^{-1}): 3429, 3332, 3221, 2226, 1628, 1554, 1487, 1259, 1081, 852, 790; ^1H NMR (DMSO, 500 MHz) (δ, ppm): 7.45-7.52 (m, 3H, aromatic H), 7.55 (d, *J* = 8.1 Hz, 2H, aromatic H), 7.62 (d, *J* = 8.1 Hz, 2H, aromatic H), 7.64 (d, *J* = 8.2 Hz, 2H, aromatic H) 7.85 (br., s, 2H, NH_2); ^{13}C NMR (DMSO, 125 MHz) (δ, ppm): 167.10, 160.24, 158.21, 133.98, 132.56, 131.01, 130.12, 129.02, 128.12, 126.45, 117.89, 116.09, 114.93, 94.29, 88.20; GC/MS: 363.07 (M^+).

2-amino-6-(phenylthio)-4-(thiophen-2-yl)pyridine-3,5-dicarbonitrile (4j): IR (KBr) (ν_{max}/cm^{-1}): 3456, 3398, 3211, 2982, 2210, 1617, 1512, 1267, 1064, 732; ^1H NMR (DMSO, 500 MHz) (δ, ppm): 6.65 (t, *J* = 4.2 Hz, 1H, aromatic H), 7.27 (d, *J* = 4.2 Hz, 1H, aromatic H), 7.35-7.40 (m, 3H, aromatic H), 7.43-7.48 (m, 2H, aromatic H), 7.72 (br., s, 2H, NH_2), 7.95 (d, *J* = 4.8 Hz, 1H, aromatic H); ^{13}C NMR (DMSO, 125 MHz) (δ, ppm): 170.11, 161.47, 146.18, 145.68, 144.14, 136.50, 130.98, 129.51, 127.86, 117.52, 116.32, 115.59, 113.80, 92.88, 83.11; GC/MS: 335.07 (M^+).

2-amino-4-(furan-2-yl)-6-(phenylthio)pyridine-3,5-dicarbonitrile (4k): IR (KBr) (ν_{max}/cm^{-1}): 3375, 3311, 3212, 2958, 2215, 1648, 1512, 1262, 1023, 764; ^1H NMR (DMSO, 500 MHz) (δ, ppm): 7.28 (t, *J* = 4.1 Hz, 1H, aromatic H), 7.45-7.50 (m, 3H, aromatic H), 7.55-7.60 (m, 3H, aromatic H), 7.80 (br., s, 2H, NH_2), 7.95 (d, *J* = 5.2 Hz, 1H, aromatic H); ^{13}C NMR (DMSO, 125 MHz) (δ, ppm): 169.23, 160.12, 159.59, 157.25, 138.36, 136.60, 132.58, 130.99, 129.56, 127.14, 120.21, 115.68, 114.42, 100.26, 88.96; GC/MS: 319.09 (M^+).

RESULT AND DISCUSSION

To begin our study, the Fe_3O_4@GO-Pr-SO_3H catalyst was prepared according to the literature procedure[xxxii] and characterized by FT-IR, X-ray diffraction (XRD), and pH analysis as reported in our previous work[xxviii]. The catalytic activity of this material was evaluated in the synthesis of pyridine dicarbonitrile derivatives. Synthesis of compound **4b** was selected as a model reaction for optimizing the reaction conditions. The reaction was carried out with 4-methyl thiophenol **1b**, malononitrile **2**, and 4-chlorobenzaldehyde **3b** in the presence of different amounts of catalyst and in various solvents and also under solvent-free conditions (Table 1). Long reaction times and poor yields of the product **4b** were obtained in the absence of the catalyst in all cases (entries 1-5). Also, low yields of the desired product were obtained under solvent-free conditions in the presence or absence of the catalyst (entries 4, 5). The presence of temperature was necessary for all situations. Therefore, the best results were reached under catalytic condition upon refluxing solvents, preferably ethanol (entries 12-16). According to the final outcomes, the reaction was more facile and proceeded to give the highest yield (95%), and short reaction time (25 min), using 0.06 g of the catalyst in ethanol (5 ml) at reflux temperature (entry 12). Increase of the

catalyst amount up to 0.08 g did not improve the product yield or shorten reaction time (entry 12).

Table 1. Optimization of reaction conditions for the synthesis of compound **4b** catalyzed by FGOSA.*

Entry	Catalyst (g)	Solvent	T/°C	Time/min	Isolated Yield/%
1	-----	H_2O	Reflux	175	32
2	-----	EtOH	Reflux	175	44
3	-----	EtOH	r.t.	175	34
4	-----	-----	100	145	16
5	-----	-----	120	145	19
6	0.06	-----	120	100	72
7	0.02	EtOH	Reflux	60	77
8	0.04	EtOH	Reflux	40	87
9	0.06	EtOH	80	45	89
10	0.06	EtOH	r.t.	30	77
11	0.08	EtOH	Reflux	20	94
12	**0.06**	**EtOH**	**Reflux**	**25**	**95**
13	0.06	H_2O	Reflux	25	83
14	0.06	MeOH	Reflux	40	63
15	0.06	CH_3CN	Reflux	50	52
16	0.06	CH_2Cl_2	Reflux	60	43

* Reaction conditions: 4-methyl thiophenol **1b**, malononitrile **2**, and 4-chlorobenzaldehyde **3b** (1.5 mmol).

According to these results, and to generalize this model reaction, we developed the reaction of malononitrile **2** with the two kinds of thiophenols **1** and various aryl aldehydes **3** under the optimized reaction conditions (Table 2). The FGOSA efficiently catalyzed the reactions, giving the desired products in high yields over relatively short reaction times. Easy separation of obtained products from the catalyst makes this method useful for the synthesis of pyridine dicarbonitriles.

Table 2. Synthesis of pyridine dicarbonitriles[a]

Entry	R	R'	Product	Time (min)	Yield (%)
1	Me	C_6H_5	**4a**	20	93
2	Me	4-Cl-C_6H_4	**4b**	25	95
3	Me	4-NO_2-C_6H_4	**4c**	21	92
4	Me	3-Br-C_6H_4	**4d**	23	90
5	Me	3-Cl-C_6H_4	**4e**	19	89
6	Me	3-NO_2-C_6H_4	**4f**	24	93
7	Me	4-CH_3O-C_6H_4	**4g**	26	91
8	Me	4-Br-C_6H_4	**4h**	27	92
9	H	4-Cl-C_6H_4	**4i**	25	89
10	H	2-thienyl	**4j**	23	90
11	H	2-furyl	**4k**	24	92

[a]Reaction conditions: thiophenols, malononitrile, aryl aldehydes, and FGOSA (0.06 g) in refluxing ethanol.

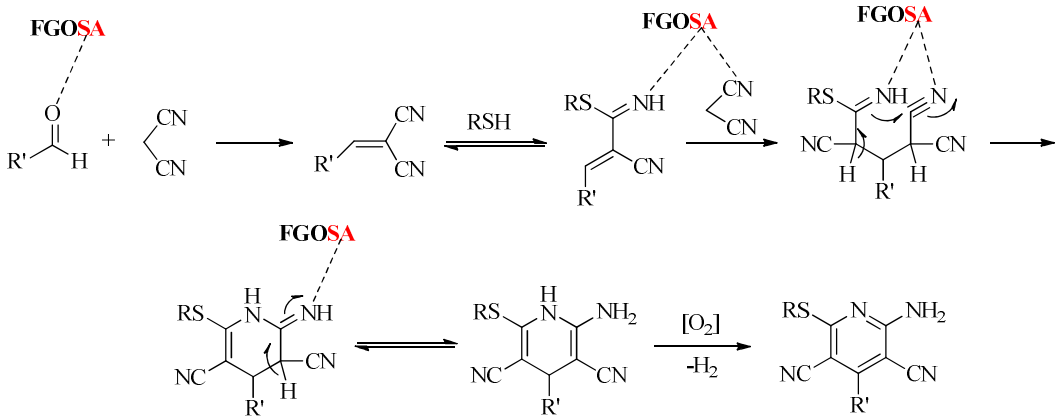
We compared the results we obtained using FGOSA as catalyst with previously reported results for the synthesis of pyridine dicarbonitriles in the presence of various catalysts (Table 3). Our reaction conditions showed shorter reaction times than all the other conditions and gave high yields of the desired products.

Table 3. Comparison of the efficiencies of different catalysts for the synthesis of pyridine dicarbonitriles.

Catalyst	Conditions		Time (min)	Yield (%)	Ref.
	Solvent	T/ºC			
Et_3N	EtOH	reflux	120	20-48	v
IBX	H_2O	70	90-150	69-83	vi
DABCO	EtOH	reflux	40-90	39-96	vii
K_2CO_3	EtOH-H_2O	reflux	40-180	60-90	viii
KOH	EtOH	reflux	30-90	71-90	ix
MS 4A	H_2O	reflux	30-120	78-91	x
Nano-CuI	EtOH-H_2O	reflux	85-200	70-94	xi
Nano-MgO	EtOH	reflux	120-540	41-69	xii
FGOSA	EtOH	reflux	19-27	89-95	This work

The model reaction under optimized reaction conditions was used for evaluating reusability of FGOSA catalyst. Upon completion of the reaction, the catalyst was recovered as described in the experimental section. The catalyst could be used at least four times without significant reduction in its activity (95, 95, 94, 94%).

Probably, the catalyst could act as a Brønsted acid and therefore promote the reactions. The catalyst would play a significant role in increasing the electrophilic character of the electrophiles in the reaction.



Scheme 2. Plausible mechanism for the FGOSA catalyzed formation of pyridine dicarbonitriles.

CONCLUSION

In this paper we developed the synthesis of pyridine dicarbonitrile derivatives in the presence of FGOSA as a highly effective heterogeneous catalyst. This method provided these products in high yields over short reaction time. Also, easy magnetic separation makes this catalyst attractive in view of green chemistry and catalysis science.

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