### FULL PAPER



# Gallic acid grafted to amine-functionalized magnetic nanoparticles as a proficient catalyst for environmentally friendly synthesis of $\alpha$ -aminonitriles

Ali Maleki ២ | Sepide Azadegan | Jamal Rahimi

Catalysts and Organic Synthesis Research Laboratory, Department of Chemistry, Iran University of Science and Technology, Tehran 16846-13114, Iran

#### Correspondence

Ali Maleki, Catalysts and Organic Synthesis Research Laboratory, Department of Chemistry, Iran University of Science and Technology, Tehran 16846-13114, Iran. Email: maleki@iust.ac.ir

**Funding information** 

Research Council of the Iran University of Science and Technology

An effective approach of one-pot catalytic Strecker reaction between aromatic aldehydes, aniline or toluidine and trimethylsilyl cyanide in the presence of amine-functionalized  $Fe_3O_4@SiO_2$  nanoparticles grafted with gallic acid (GA) as a powerful catalyst was developed. The fabricated reusable catalyst demonstrated high efficiency in the synthesis of  $\alpha$ -aminonitriles along with facile work-up procedure.  $Fe_3O_4@SiO_2$ -NH<sub>2</sub>-GA was characterized by Fourier transform-infrared spectroscopy, scanning electron microscopy image, vibrating-sample magnetometer curve, energy-dispersive X-ray analysis and thermogravimetric analysis.

### KEYWORDS

aminonitrile,  $\rm Fe_3O_4/SiO_2$  nanocomposite, gallic acid, heterogeneous green nanocatalyst, multi-component reaction

# **1** | INTRODUCTION

Multi-component reaction (MCR) processes are of great interest, not only because of their atom economy but also for their application in diversity-oriented synthesis and in preparing libraries for the screening of functional molecules.<sup>[1]</sup> The Strecker reaction, for example, is known as a significant, practical, manageable and economic tool wide-spreading usage in synthesis with of αaminonitriles. Intermediates of this useful MCR are successfully used also in the synthesis of abundant pharmaceutically important indole alkaloids, like hirsutine,<sup>[2]</sup> or eburnamonine,<sup>[3]</sup> which can have a role as potent antihypertensive agents. Regarding the various usages of  $\alpha$ aminonitriles in preparing wide-varied amino acids, amides, diamines and nitrogen-containing heterocycles, profound impact of these important substances can be recognized.<sup>[4-6]</sup> An innumerable number of acidcatalyzed Strecker reactions, including homogeneous Lewis acids,<sup>[7–9]</sup> heterogeneous Lewis acids<sup>[10,11]</sup> and also heterogeneous Brønsted acids<sup>[12,13]</sup> were reported.

It is widely acknowledged that there is a growing need for more environmentally acceptable processes and catalysts in the chemical industry. This trend has become known as green chemistry or sustainable technology.<sup>[14,15]</sup> Surface functionalization of magnetic nanoparticles (MNPs) is a well-designed way to bridge the gap between heterogeneous and homogeneous catalysis. The introduction of MNPs in a variety of solid matrices allows the combination of well-known procedures for catalyst heterogenization with techniques for magnetic separation.<sup>[16–18]</sup> So, these types of catalysts play a vital role in approaching the green chemistry goal. Beside other catalysts, MNP catalysts show excellent function in synthesis of  $\alpha$ -aminonitriles as well.<sup>[19,20]</sup>

Plenty of successful examples of silica-coated MNPs in different synthesis areas such as carbon–carbon coupling,<sup>[21,22]</sup> acetylation,<sup>[23]</sup> oxidation,<sup>[24]</sup> hydrogenation<sup>[25]</sup> and asymmetric synthesis<sup>[26]</sup> are reported.

These powerful silica-based magnetically recoverable nanocatalysts have the following attractive features: (1) good dispersion of the active species (usually noble metal 2 of 7 WILEY Organometallic Chemistry

NPs) on the magnetic support; (2) sufficient binding strength between the active species and magnetic support to enable recycling; (3) good chemical inertness of the magnetic support; (4) high saturation magnetization to facilitate magnetic separation; (5) soft ferromagnetism for redispersion; (6) a wide variety of silylating agents can be used, allowing pendant functional groups in the inorganic framework; (7) attachment is easier on silica surfaces than on organic polymeric supports (which have a high number of cross-linking bonds); (8) low cost, ready availability, mechanical robustness and straightforward synthesis; and (9) a quasi-homogeneous state can be achieved.<sup>[14]</sup>

Gallic acid (GA) is a yellowish white crystal having a molecular mass of  $170.12 \text{ g mol}^{-1}$ , melting point  $250^{\circ}$ C and water solubility 1.1% at  $20^{\circ}$ C.<sup>[27]</sup> It is a natural plant triphenol, and its derivatives have been extensively evaluated for their antitumor activity against a variety of cell lineages. Some GA derivatives such as methyl, propyl, octyl and dodecyl gallates are widely used in food manufacturing as antioxidants, as well as in the pharmaceutical and cosmetic industries. Previous studies have shown that these compounds have potent therapeutic properties, including antitumoral, antimicrobial and antiviral, as well as being potent antioxidants, acting as scavengers of reactive oxygen species.<sup>[28–31]</sup>

Herein, design and development of aminefunctionalized silica-based MNPs grafting with GA as an effective recoverable catalyst was reported for the Strecker MCR to afford  $\alpha$ -aminonitriles **4a–y** starting from an aromatic aldehyde, aniline or toluidine and



SCHEME 1 Nano-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub>@GA-catalysed protocol

trimethylsilyl cyanide (TMSCN) in excellent yields (Scheme 1). With the aim of a green approach to  $\alpha$ -aminonitriles, this is the first report on the application of Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub>-GA in this procedure.

### 2 | EXPERIMENTAL

### 2.1 | Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub>-GA preparation

Initially, 1.0 g of freshly prepared  $Fe_3O_4@SiO_2-NH_2^{[32]}$  was dispersed in a minimum amount of dry (anhydrous) toluene. A homogenous mixture of 0.6 g GA in dry toluene should be added drop-wise to the dispersed  $Fe_3O_4@SiO_2-NH_2$  at room temperature and under mild stirring conditions. Finally, 10 mol% of  $ZrCl_4$  is also needed as co-catalyst. Next, refluxing of the aforementioned mixture for 24 hr gave the desired nanocatalyst. Eventually, washing and drying resulted in the  $Fe_3O_4@SiO_2-NH_2-GA$  being applicable as a proper heterogeneous nanocatalyst.

### 2.2 | Typical procedure for the synthesis of α-aminonitrile derivatives via multi-component reaction

A mixture of an aromatic aldehyde (1 mmol), aniline or toluidine (1 mmol) and TMSCN (1 mmol) in ethanol (5 mL) in the presence of 40 mg of  $Fe_3O_4@SiO_2-NH_2-GA$  was stirred at room temperature. After completion of the reaction, which was indicated by thin-layer chromatography (TLC), the MNP catalyst was separated magnetically and the desired products were achieved by crystallization from ethanol.



**FIGURE 1** Comparative Fourier transform-infrared (FT-IR) spectra of nano-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub>@GA and previous catalyst manufacturing steps

#### MALEKI ET AL

# **3** | **RESULTS AND DISCUSSION**

# 3.1 | Characterization of the catalyst

### 3.1.1 | Fourier transform-infrared spectra

The comparative Fourier transform-infrared (FT-IR) spectra of the synthesized catalyst with the previous manufacturing levels are presented (Figure 1). Clearly, vibration peaks of Fe-O, Si-OH and Si-O-Si were associated with related ones in the other spectrums. The absorption bands at 1383 and 1657 cm<sup>-1</sup> were related to C-C stretching and C-H vibrations, respectively,<sup>[33]</sup> and the existence of an aromatic ring in the synthesized nanocatalyst could be ascertained. Finally, bands that appeared at 1504 cm<sup>-1</sup> and were attributed to the N-H and C=O at 1680 cm<sup>-1</sup> approved the successful fabrication.

# 3.1.2 | Energy-dispersive X-ray analysis

The components of the synthesized catalyst were confirmed using energy-dispersive X-ray (EDX) analysis (Figure 2). As shown in the figure, the presence of Si,



 $\label{eq:FIGURE 2} FIGURE \ 2 \quad \mbox{Energy-dispersive X-ray (EDX) analysis of (a) nano-Fe_3O_4@SiO_2-NH_2 and (b) nano-Fe_3O_4@SiO_2-NH_2@GA$ 

O, N and Fe signals implied that the silica layer was coated on the surface of the Fe<sub>3</sub>O<sub>4</sub> NPs and N linkages were formed satisfyingly. As it can be seen, the weight percentage (W%) of carbon and oxygen elements have been increased after GA loading onto the surface of the MNPs (Figure 2a and b).

# 3.1.3 | Morphology and particle size of nano-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub>@GA

The corresponding scanning electron microscopy (SEM) images of  $Fe_3O_4@SiO_2-NH_2@GA$  revealed that spherical



**FIGURE 3** (a,b) Scanning electron microscopy (SEM) images of nano-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub>@GA and (c) related particle size distribution diagram

4 of 7 WILEY-Organometallic

particles of the obtained catalyst located monotonously and no agglomeration occurred during the process (Figure 3a and b). Furthermore, the calculated average particle diameter was about 20 nm from the diameters of 52 particles (Figure 3c).

# 3.1.4 | Vibrating-sample magnetometer study

Figure 4 shows the hysteresis loops as a function of the magnetic field, or the M vs. H curve related to Fe<sub>3</sub>O<sub>4</sub>, Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>, Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub> and Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>@NH<sub>2</sub>-GA nanocomposite. The saturation magnetizations (Ms) Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>, Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub> of Fe<sub>3</sub>O<sub>4</sub>, and Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>@NH<sub>2</sub>-GA were measured as 61.50, 40.28, 21.30 and 19.1 emu  $g^{-1}$ , respectively. The decrease in saturation magnetization was most likely due to the existence of coated layers on the surface of the MNPs, which formed during the nanocatalyst manufacturing process.<sup>[34]</sup> The high resulting magnetic saturation allows easy separation of the nanocatalyst from the reaction media within a few seconds. The low coercivity of 8.64 for Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>@NH<sub>2</sub>-GA confirmed Oe the superparamagnetic property of the nanocomposite.

### 3.1.5 | Thermogravimetric analysis

Figure 5 illustrates the thermogravimetric analysis (TGA) curve of GA nanocatalyst. Weight loss at temperatures below 200°C can be attributed to water desorption from the surface of the silica layer, while weight loss above 600°C is associated with the release of the structure water.<sup>[35]</sup> Mass loss at temperature ~210–300°C corresponding to GA and another at ~400–600°C is due to the organic part of 3-aminopropylsilica.



FIGURE 4 Magnetization curve of nano-Fe $_3O_4$ @SiO $_2$ -NH $_2$ -GA in comparison with its comparative components



FIGURE 5 Thermogravimetric analysis (TGA) curve of nano-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub>-GA

**TABLE 1** Screening of reaction condition and catalyst efficiency investigation

Entry	Catalyst	Temp. (°C)	Time (min)	Solvent	Yield (%)
1	Fe <sub>3</sub> O <sub>4</sub>	r.t.	10	EtOH	65
2	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub>	r.t.	10	EtOH	78
3	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub>	r.t.	10	EtOH	80
4	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA	Reflux	10	EtOH	99
5	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA	r.t.	10	EtOH	98
6	Silica-based ionic liquid <sup>[36]</sup>	r.t.	75	EtOH	90
7	Cerium (III) chloride <sup>[37]</sup>	r.t.	85	CH <sub>3</sub> N	95
8	L-Proline <sup>[38]</sup>	r.t.	120	$\mathrm{CH}_3\mathrm{N}$	95
9	MgI <sub>2</sub> <sup>[39]</sup>	r.t.	180	$CH_2Cl_2$	97
10	-	r.t.	10	EtOH	Trace
11	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA	r.t.	20	${\rm H_2O}$	65
12	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA	r.t.	10	$\mathrm{CH}_3\mathrm{N}$	90
13	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA	r.t.	20	DMSO	68
14	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA	r.t.	20	$CH_2Cl_2$	46
15	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA (10 mg)	r.t.	10	EtOH	60
16	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA (20 mg)	r.t.	10	EtOH	84
17	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA (30 mg)	r.t.	10	EtOH	90
18	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA (40 mg)	r.t.	10	EtOH	98
19	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA (50 mg)	r.t.	10	EtOH	98
20	Fe <sub>3</sub> O <sub>4</sub> @SiO <sub>2</sub> -NH <sub>2</sub> -GA (60 mg)	r.t.	10	EtOH	98
21	GA	r.t.	10	EtOH	56

GA, gallic acid.

### VILEY Chemistry

# 3.1.6 | Monitoring the reaction conditions

Referring to the gathered information in hand from various analyses, the claim of catalyst fabrication can be proved. The feasibility of the suggested strategy and optimizing reaction conditions were probed on the reaction of benzaldehyde, aniline and TMSCN as a pattern reaction. Furthermore, the nanocatalyst was assessed as a powerful catalyst for synthesis of  $\alpha$ -aminonitrile derivatives. To achieve the mentioned goal, the efficiency of the introduced nanocatalyst was also compared with several catalysts. The pattern reaction was carried out with the best yield using Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>-NH<sub>2</sub>-GA as a catalyst in comparison with gained catalysts from previous fabrication levels (Table 1, entries 1–4; see also results of some reported procedures for further comparison – entries 6–

9). Solvent screening revealed that the reaction performed well in EtOH compared with other suggested solvents with different polarities under identical conditions (entries 5 and 11–14). To find the best reaction conditions, a set of experiments was carried out by changing the catalyst amount. According to the results, 40 mg of catalyst is sufficient to promote the reaction with satisfactory yield (entries 15–21).

# 3.1.7 | Investigation of catalyst activity

The obtained results provided an incentive to extend the recommended process to various aromatic aldehyde substrates. The corresponding  $\alpha$ -aminonitriles were obtained in 85–98% yields (Table 2). Regarding the eco-friendly

TABLE 2	GA-grafted amine-functionalized MNPs-catalyzed synthesis of $\alpha$ -aminonitriles

						M.P. (°C)	
Entry	$\mathbb{R}^1$	$\mathbb{R}^2$	Product	Time (min)	Yield <sup>a</sup> (%)	Obs.	Lit.
1	Phenyl	Н	4a	10	98	80-83	73-75 <sup>[40]</sup>
2	4-Chlorophenyl	Н	4b	10	97	113-115	111-112 <sup>[33]</sup>
3	4-Bromophenyl	Н	4c	5	98	102-106	$100 - 101^{[40]}$
4	3-Nitrophenyl	Н	4d	10	94	84-88	89-90 <sup>[40]</sup>
5	4-Methoxylphenyl	Н	4e	15	92	91–92	91-92 <sup>[40]</sup>
6	4-Nitrophenyl	Н	4f	5	98	86-88	86-88 <sup>[40]</sup>
7	4-Hydroxyphenyl	Н	4g	15	93	124-126	124-125 <sup>[40]</sup>
8	4-Methylphenyl	Н	4h	20	92	71–73	70-71 <sup>[40]</sup>
9	4-Fluorophenyl	Н	<b>4i</b>	10	97	101-102	99–101 <sup>[34]</sup>
10	2-Thienyl–	Н	4j	20	88	97–99	97–99 <sup>[19]</sup>
11	2-Furanyl	Н	4k	20	90	73–75	72–74 <sup>[19]</sup>
12	4-(Dimethylamino)phenyl	Н	41	25	90	107-109	107-110 <sup>[19]</sup>
13	4-Cyanophenyl	Н	4m	10	98	112-114	113-115 <sup>[35]</sup>
14	3,4,5-Trimethoxyphenyl	Н	4n	10	95	146-148	147-149 <sup>[34]</sup>
15	4-Hydroxy-3-methoxyphenyl	Н	40	20	87	114–116	113-115 <sup>[31]</sup>
16	2-Nitrophenyl	Н	4p	15	89	135–137	135-137 <sup>[40]</sup>
17	2,6-Dichlorophenyl	Н	4q	20	88	74–76	75-78 <sup>[19]</sup>
18	Cinnamaldehyde	Н	4r	20	95	121-123	$117 - 118^{[12]}$
19	2-Chlorophenyl	Н	4s	25	85	69–71	69-70 <sup>[35]</sup>
20	3-Fluorophenyl	Н	4t	5	91	91–93	91-92 <sup>[41]</sup>
21	Phenyl	Me	4u	5	98	106-108	105-107 <sup>[35]</sup>
22	4-Chlorophenyl	Me	4v	5	98	87-89	85-87 <sup>[34]</sup>
23	Cinnamaldehyde	Me	<b>4</b> w	10	96	104-106	103-106 <sup>[34]</sup>
24	4-Methylphenyl	Me	4x	10	91	116-118	116-118 <sup>[31]</sup>
25	4-Nitrophenyl	Me	4y	5	98	84-86	83-85 <sup>[19]</sup>

<sup>a</sup>Isolated yield.



SCHEME 2 Proposed reaction cycle of synthesis of  $\alpha$ -aminonitriles in the presence of the nanocatalyst

aspects of the present work, including nanomagnetic heterogeneous catalyst (according to facile handling and purify through an external magnet along with ease of recoverable property), green solvent and room temperature conditions, the suggested protocol could be considered as environmentally friendly synthesis of  $\alpha$ -aminonitriles.

### 3.1.8 | Mechanistic evaluation

A proposed mechanism for the formation of  $\alpha$ aminonitrile derivatives is shown in Scheme 2. Through activating the oxygen atom of the carbonyl by Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>@NH<sub>2</sub>-GA catalyst, intermediate I was formed. In the case of iminium intermediate II, it is reasonable to assume that it can be a consequence of the condensation reaction between intermediate I and





different amines **2** in the presence of the catalyst. The following nucleophilic attack of TMSCN to imine yielded the products **4a–y**.

### 3.1.9 | $Fe_3O_4$ (SiO<sub>2</sub>-NH<sub>2</sub>-GA reusability

Encouraged by the mentioned analyses results, the catalyst reusability was also studied. Upon completion of the pattern reaction (monitored by TLC), the catalyst was separated using an external magnet. The recovered catalyst was then added to a fresh reaction mixture under identical conditions. The GA nanocatalyst could be efficiently recovered up to eight times without suffering any significant losses in its catalytic activity or the yield of the model reaction (Figure 6). Also, the reaction was repeated using acid-washed catalyst and it was observed that the reaction yield increased (entry 8). Therefore, some deactivated hydroxyl groups of GA were activated again by washing the catalyst with acid (HCl, 0.5 M).

# 4 | CONCLUSIONS

A new facet of the synthesis and operational nano-Fe<sub>3-</sub> $O_4$ @SiO<sub>2</sub>-NH<sub>2</sub>-GA usage in achievements of the Strecker reaction was accomplished in the present work. Furthermore, the most interesting features of the suggested procedure could be named as one-pot MCR, excellent yield,

brief time period of reaction, green solvent, facile workup procedure, ease of separation, and recyclability of the magnetic catalyst.

### ACKNOWLEDGEMENTS

The authors gratefully acknowledge the financial support from the Research Council of the Iran University of Science and Technology.

### ORCID

Ali Maleki 🕩 https://orcid.org/0000-0001-5490-3350

### REFERENCES

- A. Maleki, J. Rahimi, O. M. Demchuk, A. Z. Wilczewska, R. Jasiński, Ultrason. Sonochem, 2018, 43, 262.
- [2] M. Lounasmaa, J. Miettinen, P. Hanhinen, R. Jokela, *Tetrahe*dron Lett. 1997, 38, 1455.
- [3] A. D. S. Goes, C. Ferroud, J. Santamaria, *Tetrahedron Lett.* 1995, 36, 2235.
- [4] W. L. Matier, D. A. Owens, W. T. Comer, D. Deitchman, H. C. Ferguson, R. J. Seidehamel, J. R. Young, J. Med. Chem. 1973, 16, 901.
- [5] L. M. Weinstock, P. Davis, B. Handelsman, R. J. Tull, J. Org. Chem. 1967, 32, 2823.
- [6] G. Dyker, Angew. Chem. Int. Ed. 1997, 36, 1700.
- [7] S. K. De, R. A. Gibbs, Tetrahedron Lett. 2004, 45, 7407.
- [8] A. Maleki, Tetrahedron. 2012, 68, 7827.
- [9] Z.-L. Shen, S.-J. Ji, T.-P. Loh, Tetrahedron 2008, 64, 8159.
- [10] B. Karimi, A. Maleki, Chem. Commun. 2009, 5180.
- [11] B. Karimi, A. Maleki, D. Elhamifar, J. H. Clark, A. J. Hunt, *Chem. Commun.* **2010**, *46*, 6947.
- [12] A. Shaabani, A. Maleki, M. R. Soudi, H. Mofakham, *Cat. Com.* 2009, 10, 945.
- [13] A. Shaabani, A. Maleki, Appl. Catal. A 2007, 331, 149.
- [14] A. Maleki, J. Rahimi, J. Porous Mat. 2018, 25, 1789.
- [15] A. Maleki, V. Eskandarpour, J. Rahimi, N. Hamidi, Carbohydr. Polym. 2019, 208, 251.
- [16] A. Maleki, M. Niksefat, J. Rahimi, Z. Hajizadeh, BMC Chemistry. 2019, 13, 19.
- [17] A. Maleki, N. Hamidi, S. Maleki, J. Rahimi, Appl. Organomet. Chem. 2018, 32, e4245.
- [18] M. Azizi, A. Maleki, F. Hakimpoor, R. Firouzi-Haji, M. Ghassemi, J. Rahimi, *Lett. Org. Chem.* 2018, 15, 753.
- [19] A. Maleki, J. Rahimi, Z. Hajizadeh, M. Niksefat, J. Organomet. Chem. 2019, 881, 58.

- [20] A. Maleki, E. Akhlaghi, R. Paydar, Appl. Organomet. Chem. 2016, 30, 382.
- [21] A. Maleki, R. Taheri-Ledari, M. Soroushnejad, *ChemistrySelect* **2018**, *3*, 13 057.
- [22] A. Maleki, Tetrahedron Lett. 2013, 54, 2055.
- [23] C. Ó. Dálaigh, S. A. Corr, Y. Gun'ko, S. J. Connon, Angew. Chem. 2007, 119, 4407.
- [24] A. Maleki, Ultrason. Sonochem. 2018, 40, 460.
- [25] R. B. N. Baig, R. S. Varma, ACS Sustainable Chem. Eng. 2013, 1, 805.
- [26] A. Schätz, M. Hager, O. Reiser, Adv. Funct. Mater. 2009, 19, 2109.
- [27] S. Verma, A. Singh, A. Mishra, *Environ. Toxicol. Pharmacol.* 2013, 35, 473.
- [28] M. Inoue, R. Suzuki, N. Sakaguchi, Z. Li, T. Takeda, Y. Oghara, B. Y. Jiang, Y. Chen, *Biol. Pharm. Bull.* **1995**, *18*, 1526.
- [29] Y.-Y. Ow, I. Stupans, Curr. Drug Metab. 2003, 4, 241.
- [30] L. A. Savi, P. C. Leal, T. O. Vieira, R. Rosso, R. J. Nunes, R. A. Yunes, T. B. Creczynski-Pasa, C. R. M. Barardi, C. M. O. Simões, *Arzneimittelforschung* **2005**, *55*, 66.
- [31] M. Eslami, M. G. Dekamin, L. Motlagh, A. Maleki, Green Chem. Lett. Rev. 2018, 11, 36.
- [32] A. Maleki, S. Azadegan, J. Inorg. Organomet. Polym. Mater. 2017, 27, 714.
- [33] A. Maleki, H. Movahed, R. Paydar, RSC Adv. 2016, 6, 13 657.
- [34] K. Niknam, D. Saberi, M. N. Sefat, *Tetrahedron Lett.* 2959, 2010, 51.
- [35] J. Wang, S. Zheng, Y. Shao, J. Liu, Z. Xu, D. Zhu, J. Colloid Interface Sci. 2010, 349, 293.
- [36] M. N. Sefat, D. Saberi, K. Niknam, Catal. Lett. 2011, 141, 1713.
- [37] M. A. Pasha, H. M. Nanjundaswamy, V. P. Jayashankara, Synth. Commun. 2007, 37, 4371.
- [38] A. Nasreen, Tetrahedron Lett. 2013, 54, 3797.
- [39] Y. Lu, Y. Wang, X. Zhang, J. Chem. Res. 2013, 37, 709.
- [40] A. Maleki, R. F. Haji, M. Ghassemi, H. Ghafuri, J. Chem. Sci. 2017, 129, 457.
- [41] A. Mobaraki, B. Movassagh, B. Karimi, ACS Comb. Sci. 2014, 16, 352.

How to cite this article: Maleki A, Azadegan S, Rahimi J. Gallic acid grafted to amine-functionalized magnetic nanoparticles as a proficient catalyst for environmentally friendly synthesis of  $\alpha$ -aminonitriles. *Appl Organometal Chem.* 2019;e4810. https://doi.org/10.1002/aoc.4810