

Aluminum Oxide Nanoparticle as a Valuable Heterogeneous Nanocatalyst in the Synthesis of 2-Aminothiazole Scaffolds

Z. Shokrani^a, Z. Zarnegar^b and J. Safari^{a,*}

^aLaboratory of Organic Compound Research, Department of Organic Chemistry, College of Chemistry and Biochemistry, University of Kashan, P. O. Box: 87317-51167, Kashan, I.R, Iran

^bResearch Affiliate, Department of Chemistry, University of Payame Noor, P. O. Box: 19395-4697, Tehran, Iran

(Received 30 November 2018, Accepted 8 April 2019)

An efficient methodology for the preparation of 2-aminothiazoles is achieved by one-pot reaction of methylcarbonyls, thiourea and iodine in the presence of aluminum oxide nanoparticles as an active nanocatalyst at 85 °C in DMSO solvent. Al₂O₃ nanoparticles have been prepared *via* co-precipitation method and their structures were confirmed using Fourier transform infrared radiation (FT-TR), X-ray diffraction (XRD), scanning electron microscopic (SEM), and energy-dispersive analysis of X-ray (EDAX) analyses. Advantages of this efficient synthetic approach include higher purity and excellent yield of products, easy isolation of products and convenient manipulation.

Keywords: 1,3-Thiazoles, Al₂O₃ nanoparticles, Methylcarbonyls, One-pot synthesis, Heterogeneous catalyst

INTRODUCTION

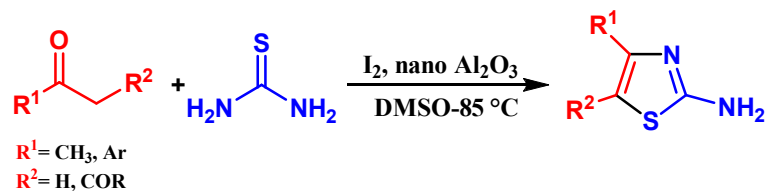
Thiazoles or 1,3-thiazoles are members of the azole <https://en.wikipedia.org/wiki/Azoleheterocycles> that contain both sulfur and nitrogen in their aromatic rings. Thiazole and their substituted analogues are very important privileged scaffolds in pharmacological and biological molecules, as building blocks for organic synthesis, natural products, and synthetic drugs [1,2]. Synthesis of 1,3-thiazoles, with pharmacological and biological properties has widely been explored by pharmacists and chemistry researchers [3,4]. The first report for the synthesis of 1,3-thiazole is proposed by Hantzsch and Weber in 1887. The Hantzsch thiazole synthesis is a reaction between haloketone and thioamides or thiourea [5]. Among the important thiazoles heterocycles, 2-aminothiazole and its derivatives have attracted increasing attention from synthetic chemists due to their diverse biological activities such as antidiabetic [6], anticancer and antitumor activity [7], antiviral agent [8], antihypertensive activity [9], antimicrobial activities [10], anti-inflammatory [11],

antileishmanial [12], anticonvulsant activity [13], *etc.* [14]. Moreover, various types of catalysts have been used such as KI/NH₄NO₃ [15], NEt₃ [16], TSOH [17], montmorillonite [18], NaCl₂ [19], (bmim)BF₄ [20], asparagine [21], silica chloride [22], 1,3-di-*n*-butylimidazolium tetrafluoroborate [23], and ammonium 1,2-molybdophosphate [24]. Despite the merits of using the methods mentioned, they include some disadvantages such as the use of expensive or toxic catalysts, harsh reaction conditions, poor atom-efficiency, long reaction time, unsatisfactory yields, complex work-up, and poor procedure.

Due to the importance of 2-aminothiazole scaffolds and fixing the limitations of reported protocols, there is still the need to develop environmentally friendly techniques and the use of effective nanocatalysts for the synthesis of these heterocycles.

In the past decades, the research on the application of metal oxide nanoparticles as advanced nanocatalysts has attracted significant attention for a wide variety of catalytic processes due to many benefits such as their high chemical activity, large surface-to-volume ratio, specificity of interaction, low coordinating sites, and easy separation from the reaction environment. Moreover, they can be applied as

*Corresponding author. E-mail: safari@kashanu.ac.ir

Scheme 1. The synthesis of 2-aminothiazole using nano γ - Al_2O_3

heterogeneous catalytic systems with the efficiency and advantage of homogenous catalysts. When the particle size decreases to nano levels and the surface area of the catalyst increases tremendously, the nanocatalyst will have a better performance in the highest catalytic activity and the more desirable potential for improving the efficiency, selectivity, and yield of catalytic processes [25-27].

Alumina (Al_2O_3) as an important family of metal oxides are utilized for diverse applications in industry and chemistry, such as catalysis in petroleum refining, electronics, ceramic composites, optoelectronics, wear protection, metallurgy, refractories, automotive emission control, and hydrogenation [28-33]. Alpha (α), beta (β) and gamma (γ) are different phases of alumina. Among the transitional alumina, γ - Al_2O_3 is the most employed one for applications in catalysis and adsorption, because of its high surface area and good porosity parameters [34]. On the other hand, nano γ - Al_2O_3 as a catalytic support and nanocatalyst has been explored for organic conversions [34-37]. In practical applications, the use of γ - Al_2O_3 nanoparticles led to the development of their catalytic activities due to some of advantages for example large surface area, pore volume, pore-size distribution, and acid/base characteristics [36,37].

With regard to the above facts, herein we would like to report nano Al_2O_3 as a valuable nanocatalyst for the synthesis of 2-aminothiazoles by one pot condensation of various methylcarbonyls and thiourea as precursors using iodine as oxidizing reagent at 85 °C in DMSO (Scheme 1).

EXPERIMENTAL PROCEDURES AND MATERIALS

Chemical substances in high purity were purchased from Merck, Fluka and Aldrich with commercial grade. Melting points (°C) were determined in an open-glass capillary

using an electro-thermal digital melting point apparatus and are uncorrected. ^1H NMR and ^{13}C NMR spectra were recorded with a Bruker DRX-400 spectrometer at 400 and 100 MHz, respectively. NMR spectra were obtained in $\text{DMSO-}d_6$ solutions and are reported as parts per million (ppm) downfield from Me_4Si as internal standard. FT-IR spectra of all the compounds were measured on a Perkin-Elmer 550 spectrometer in the range 400-4000 cm^{-1} with KBr pellets. Progress of the catalytic process was monitored by thin layer chromatography (TLC) using silica gel plates in the solvent system (ethyl acetate/*n*-hexane, 4:2). Nano γ - Al_2O_3 was characterized using a Holland Philips Xpert X-ray powder diffraction (XRD) diffractometer (CuK α radiation, $\lambda = 0.154056$ nm), at a scanning speed of 2°/min from 10° to 100° (2 θ). Scanning electron microscopic (SEM) images were performed on a Zeiss EM10C SEM that it equipped with energy dispersive X-ray spectrometer (EDAX).

Preparation of the γ - Al_2O_3 Nanoparticles

0.5 g of aluminum chloride (AlCl_3) was dissolved in 150 ml of ethanol. Then, a little amount of distilled water was added to it leading to a transparent solution. In the next step, 60 ml NH_3 as a precipitant agent was added dropwise to the stirred solution until the white Al(OH)_3 was precipitated. After filtering in vacuum system, the sample was dried at 200 °C for 2 h in oven and heated at 1250 °C for 1 h to afford white γ - Al_2O_3 nanoparticles [38].

General Procedure for the Catalytic Synthesis of 2-Aminothiazoles

A mixture of methylcarbonyl (5 mmol), thiourea (7.5 mmol), I_2 (5 mmol), and nano γ - Al_2O_3 (1 g) in DMSO was stirred at 85 °C for desired time. After the completion of the reaction (monitored by TLC), the insoluble catalyst was separated by simple filtration. The reaction media (DMSO)

was quenched by the addition of 10 ml ammonia to pH = 9-10 to give the solid products. Finally, the pure product was recrystallized from ethanol in high yield. All the corresponding products are known and were characterized by comparison of their physical (Mp) and spectral data (IR and ¹H NMR) with those of authentic samples.

4-(4-Phenyl)thiazol-2-amine. Yellow crystal; IR (KBr, cm⁻¹): 3435 (NH₂), 3254 (NH₂), 3114 (aromatic C-H), 1598 (C=C), 1517 (C=N), 1337 (N-H Bending aromatic), 773 (S-C=N); ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm): δ_H 6.92 (s, 1H, thiazole), 7.22 (s, 2H, NH₂, D₂O exchangeable), 7.32 (t, *J* = 7.9 Hz, 1H, Ar-H), 7.69 (t, *J* = 7.9 Hz, 2H, Ar-H), 7.71 (m, 2H, Ar-H).

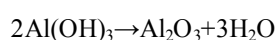
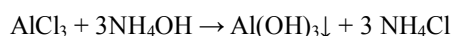
4-(4-Bromophenyl)thiazol-2-amine. Brown solid; IR (KBr, cm⁻¹): 3428 (NH₂), 3281 (NH₂), 3110 (aromatic C-H), 1632 (C=C), 1532 (C=N), 1471 (N-H Bending aromatic), 725 (S-C=N); ¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm): δ_H 7.07 (s, 1H, thiazole), 7.10 (s, 2H, NH₂), 7.53 (d, *J* = 6.8 Hz, 2H, Ar-H), 7.74 (d, *J* = 6.8 Hz, 2H, Ar-H).

4-Methyl-5(ethoxycarbonyl)thiazol-2-amine. White powder; IR (KBr, cm⁻¹): 3373 (NH₂), 3083 (aromatic C-H), 1675 (C=C), 1516 (C=N), 1373 (N-H Bending aromatic), 757 (S-C=N).

¹H NMR (DMSO-*d*₆, 400 MHz) δ (ppm): δ_H 1.22 (t, *J* = 7.1, 3H, CH₃), 2.36 (s, 3H, CH₃), 4.14 (q, 2H, CH₂), 7.72 (s, 2H, NH₂).

RESULTS AND DISCUSSION

The Al₂O₃ nanoparticles were synthesized by precipitation of aluminum trichloride in ethanol solution using as a precursor and ammonia as the precipitating agent [38]. This reaction is performed according to the following reaction [36]. The structure of γ-Al₂O₃ is identified by the characterization techniques such as FT-IR, SEM, EDAX, and XRD. Then, the catalytic behavior of γ-Al₂O₃ is evaluated for the preparation of 2-aminothiazoles.



The morphology of γ-Al₂O₃ nanoparticles was investigated by SEM technique. The SEM images, shown in Fig. 1,

indicate that Al₂O₃ nanoparticles has a nearly spherical or irregular hexagonal structure with the size distribution of 40-50 nm (Fig. 1c).

The EDAX of the γ-Al₂O₃ nanoparticles is presented in Fig. 2. These results indicate that Al₂O₃ nanoparticles have been successfully synthesized.

The FT-IR spectrum of γ-Al₂O₃ nanoparticles is shown in Fig. 3. The vibration bands at 3457 cm⁻¹ and 1640 cm⁻¹ are attributed to asymmetrical stretching of O-H and the bending vibration of adsorbed water or alcohol, respectively. The wide band appeared between 500 cm⁻¹ and 900 cm⁻¹ corresponds to the stretching vibration of co-ordinate O-Al-O bond, showing nano amorphous Al₂O₃ [38,39].

Figure 4 shows the XRD pattern of γ-Al₂O₃ nanoparticles. The characteristic XRD peak of aluminum oxide was displayed at 2θ 37.75°, 39.32°, 46.07°, 61.44° and 67.19° which are assigned to the diffractions of the (110), (111), (210), (210) and (211) reflections, respectively. These peaks are related to the Al₂O₃ nanoparticles with the crystalline cubic structure which are in accordance with aluminum oxide (JCDPS card no. 01-1303) [38].

After identifying the structure of the γ-Al₂O₃ nanoparticles, their catalytic activity was evaluated in the synthesis of thiazole derivatives. Therefore, acetophenone, thiourea, and iodine were chosen as the model substrates in DMSO solvent. The influence of quantity of nanocatalyst, and the effect of temperature and solvent were studied to optimize the reaction conditions. The obtained results are listed in Table 1. The model reaction without nanocatalyst resulted in the formation of product 4a with 75% yield for 4 h. Determining the minimum amount of catalyst is necessary for maximum formation of the desired product. As shown in this table, the reaction yield improved with increasing the amount of nanocatalyst from 0.4 to 1.1 g with shorter reaction time. When nano γ-Al₂O₃ used was 1 g, the product formation enhanced rapidly with 94% yield in 3 h. To select the appropriate temperature, the reaction was checked in the temperature range from 60-90 °C. It was observed that the optimized yield was obtained at 85 °C in DMSO. Other solvents were not effective in improving this catalytic reaction.

In order to explore the scope and generality of this methodology, different types of methylcarbonyls with

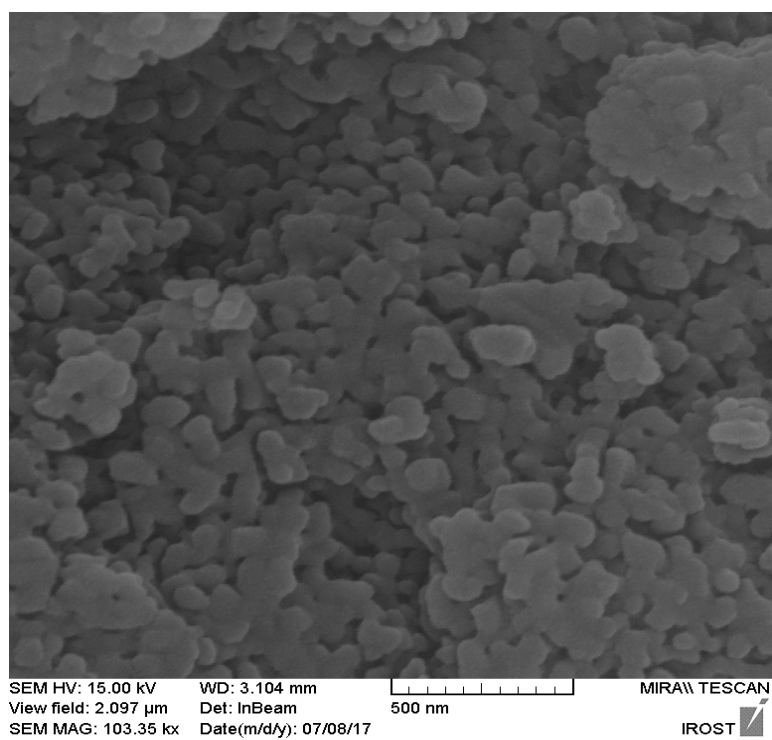
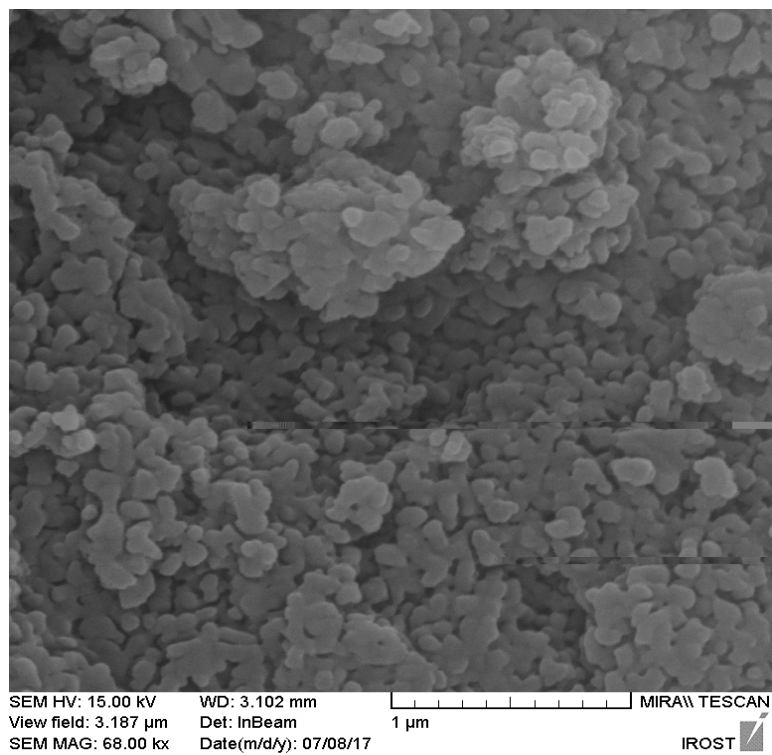


Fig. 1. (a) and (b) SEM images of $\gamma\text{-Al}_2\text{O}_3$ nanoparticles, and (c) the histogram of the particles size distribution.

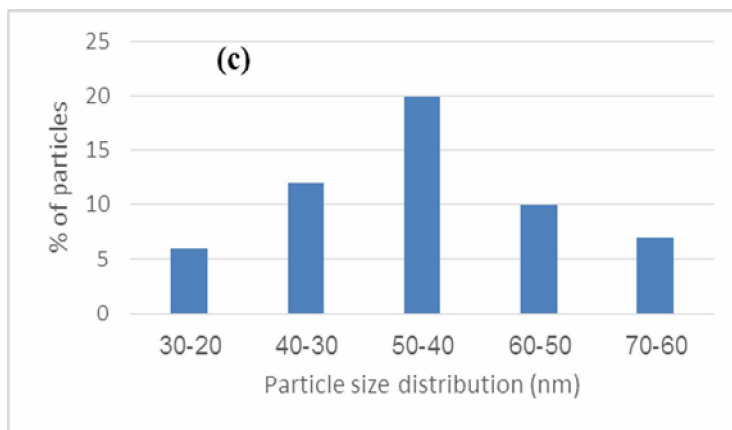


Fig. 1. Continued.

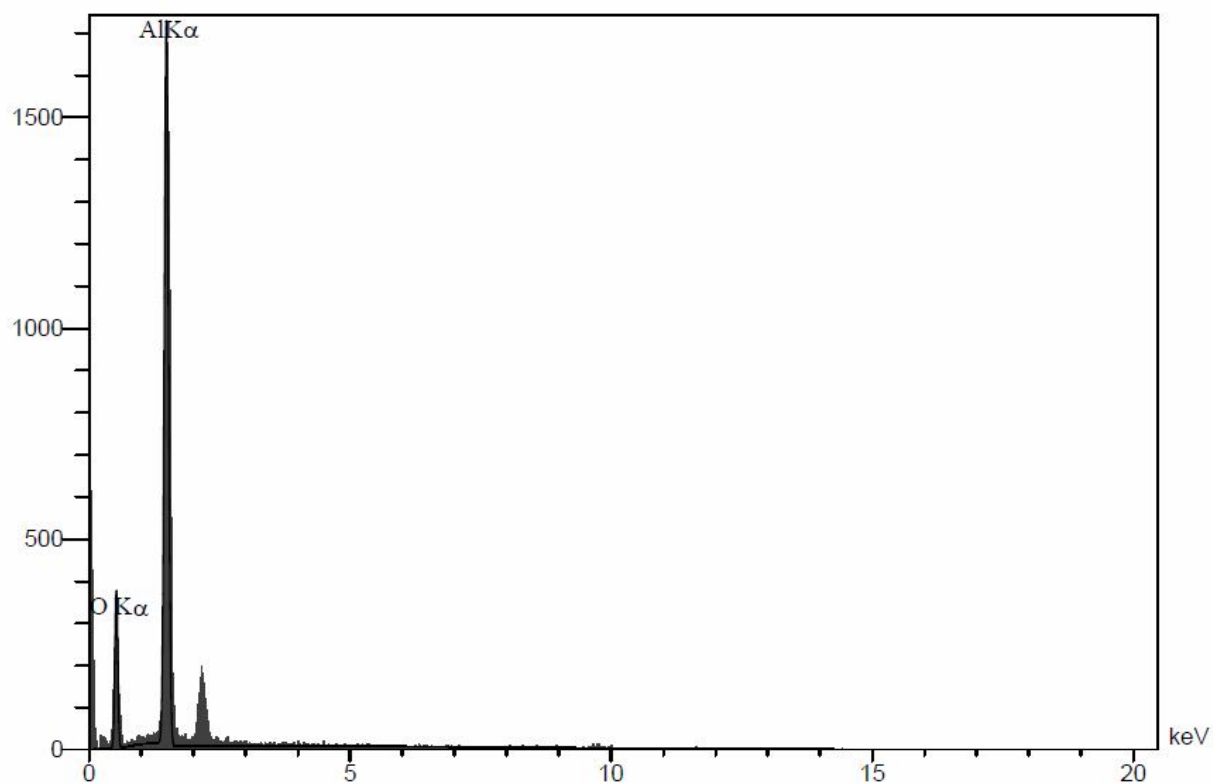


Fig. 2. EDaX analysis of γ - Al_2O_3 nanoparticles.

electron-releasing groups or electron-withdrawing were evaluated. The results are summarized in Table 2. The catalytic reaction proceeded with a variety of

methylcarbonyls in the presence of nano Al_2O_3 at 85 °C and the desired products were obtained in good to excellent yields (85-96%). The procedure was highly effective and the

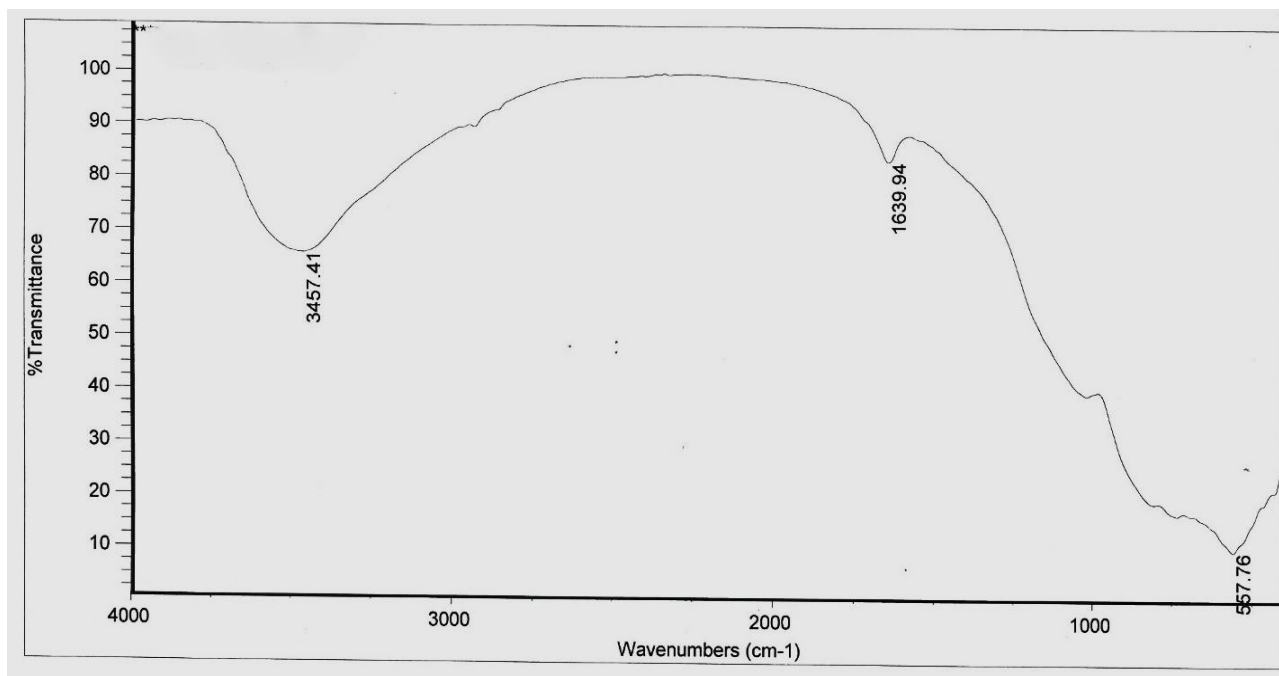


Fig. 3. FT-IR spectrum of γ -Al₂O₃ nanoparticles.

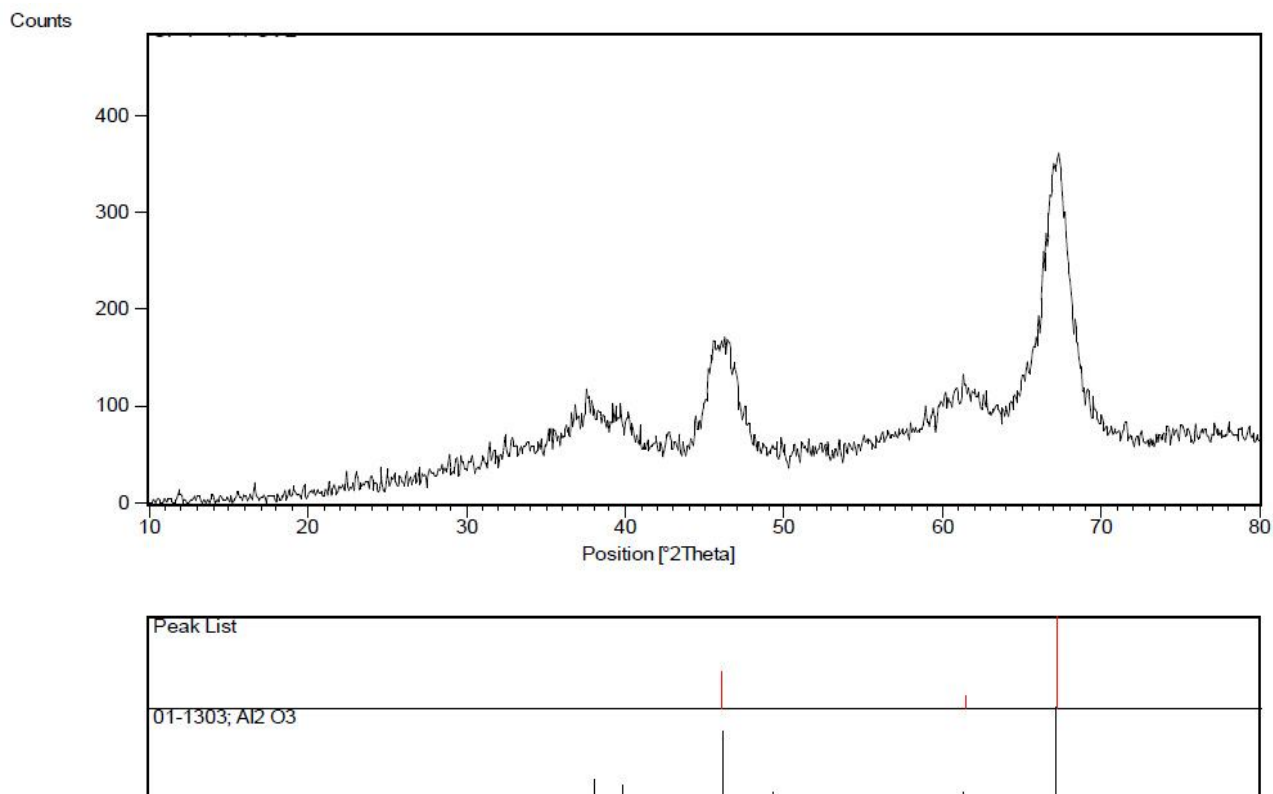
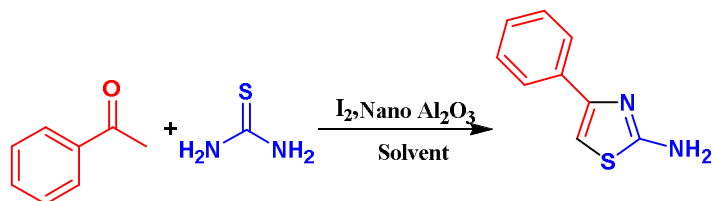


Fig. 4. XRD pattern of γ -Al₂O₃ nanoparticles.

Table 1. Optimization of Reaction Conditions of Model Cyclocondensation^a

Entry	Catalyst (g)	Solvent	Temperature (°C)	Time (h)	Yield (%) ^b
1	-	DMSO	85	4	75
2	0.4	DMSO	85	3.5	83
3	0.6	DMSO	85	3.5	87
4	0.9	DMSO	85	3	93
5	1.0	DMSO	85	3	94
6	1.1	DMSO	85	3	94
7	1.0	DMSO	60	5	84
8	1.0	DMSO	70	4	92
9	1.0	DMSO	80	3	95
10	1.0	DMSO	90	3	93
11	1.0	EtOH	reflux	3	90
12	1.0	MeOH	reflux	5	25
13	1.0	H ₂ O	reflux	5	30
14	1.0	CH ₃ CN	reflux	3	89
15	1.0	THF	reflux	4	87

^aReaction conditions: acetophenone (5 mmol), thiourea (7.5 mmol), I₂ (5 mM) and solvent (5 ml).

^bIsolated yield.

nature of substituent on the aromatic ring of aldehydes did not show obvious effects in terms of yields and times under the optimized conditions.

A mechanistic pathway for the catalytic synthesis of 2-aminothiazoles is exemplified in Scheme 2. The γ -Al₂O₃ nanoparticles facilitates this cyclocondensation through Lewis acid-Lewis basic catalysis approach. In the suggested mechanism, Al³⁺ as a Lewis acid active site polarizes the

carbonyl group of acetophenone and O²⁻ of Al₂O₃ acts as a Lewis base in the formation of enolate to produce α -iodoacetophenone. Additionally, thiourea is converted to enol under basic conditions and react with the iodinated acetophenone for the synthesis of carbamimidothioic acid species. Then, Al₂O₃-promoted dehydrogenation gives 2-aminothiazoles [40,42].

The catalytic role of Al₂O₃ for the synthesis of

Table 2. Synthesis of 2-Aminothiazoles Using Different Substrates

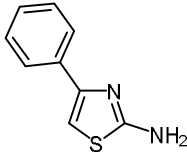
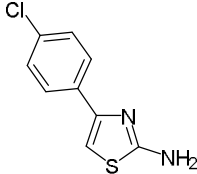
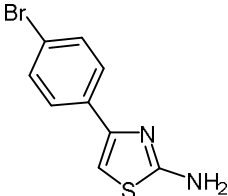
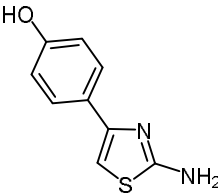
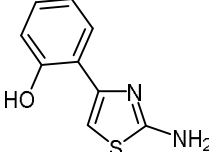
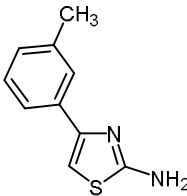
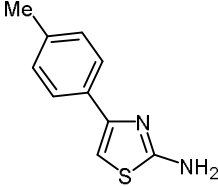
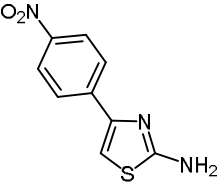
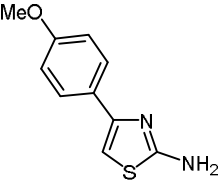
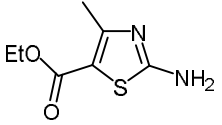
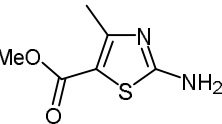
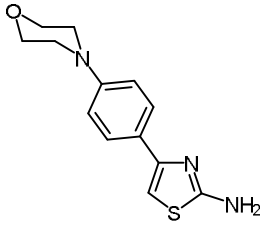
Entry	Product	Time (h)	Yield (%) ^a	M.p. (°C) (Ref.)
1		3	94	149-151 (151-153) [40]
2		3.5	96	160-162 (161-162) [40]
3		3.5	96	181-182 (180-183) [40]
4		4	92	197-199 (198-200) [40]
5		5	93	139-141 (138-139) [40]
6		3.5	92	88-90 (88-91) [40]
7		4	90	151-152 (152) [41]

Table 2. Continued

8		3	85	286-287 (285-286) [40]
9		4	87	140-142 (141) [41]
10		3	91	177-178 (177-179) [40]
11		3	88	224-226 (225-226) [40]
12		3	96	278-281 (277-280) [42]

^aIsolated yield.

2-aminothiazoles has been compared with previously reported catalysts. The results are summarized in Table 3. As indicated in this table, the catalytic performance of the Al₂O₃ nanocatalyst leads to a better catalytic activity and higher yield compared to other catalysts.

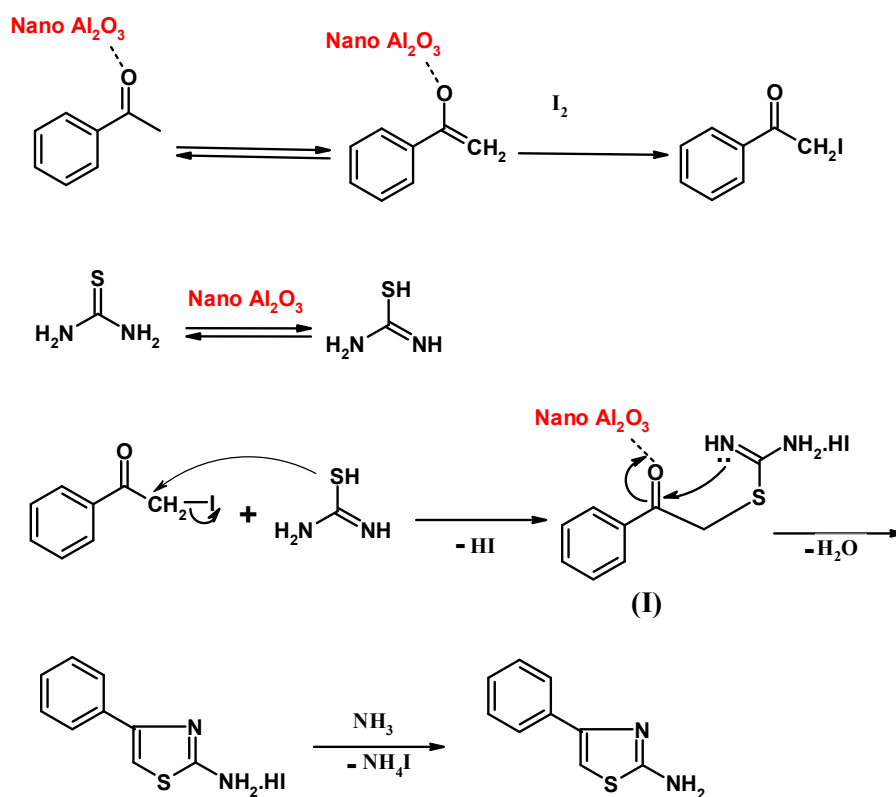
The reusability of the Al₂O₃ was evaluated using the model reaction under the optimized conditions. For this purpose, after completion of the reaction, the catalyst was retrieved from the reaction medium by simple filtration. After being washed with ethyl acetate for several times and dried at 50 °C, the catalyst can be reused without any deactivation even after five successive cycles of synthesis of thiazoles (Fig. 5).

CONCLUSIONS

In this research, we have described an efficient and simple method for the synthesis of 2-aminothiazoles catalyzed by Al₂O₃ nanoparticles. The present protocol offers some of advantages such as high yields of the products, short reaction time, and application of nanotechnology in the catalytic process.

ACKNOWLEDGMENTS

We gratefully acknowledge the financial support from the Research Council of the University of Kashan for



Scheme 2. Plausible Mechanism for the synthesis of 2-aminothiazoles

Table 3. Comparison Al_2O_3 Nanocatalyst with other Catalysts in the Literature to Synthesize 4-(4-Phenyl)thiazol-2-amine

Entry	Conditions	Time (h)	Temperature ($^{\circ}\text{C}$)	Yield (%) [Ref.]
1	$\text{Fe}_3\text{O}_4@\text{SiO}_2\text{-TiO}_2$ (10 mol%) (PEG-200)	5	r.t	98 [4]
2	TsOH (0.25 mmol), DMSO	12	80	55 [17]
3	Pyrrolidine (0.2 ml), EtOH	4	90	70 [16]
4	Diethanolamine (0.2 ml), EtOH	4	90	70 [16]
5	NEt_3 (0.2 ml), EtOH	4	80	96 [16]
6	Valine (7.5 mol%), DMSO	4	85	75 [21]
7	Asparagine (7.5 mol%), DMSO	4	85	95 [21]
8	$\text{KI}/\text{NH}_4\text{NO}_3$ (20 mol%), H_2SO_4 (1 eq), O_2 , [Bmim]OTf/ H_2O	4	40	95 [15]
9	Catalyst-free, PEG-400	6	r.t	90 [43]
10	$\gamma\text{-Al}_2\text{O}_3$ nanoparticles (1 g), DMSO	3	85	94 (This research)

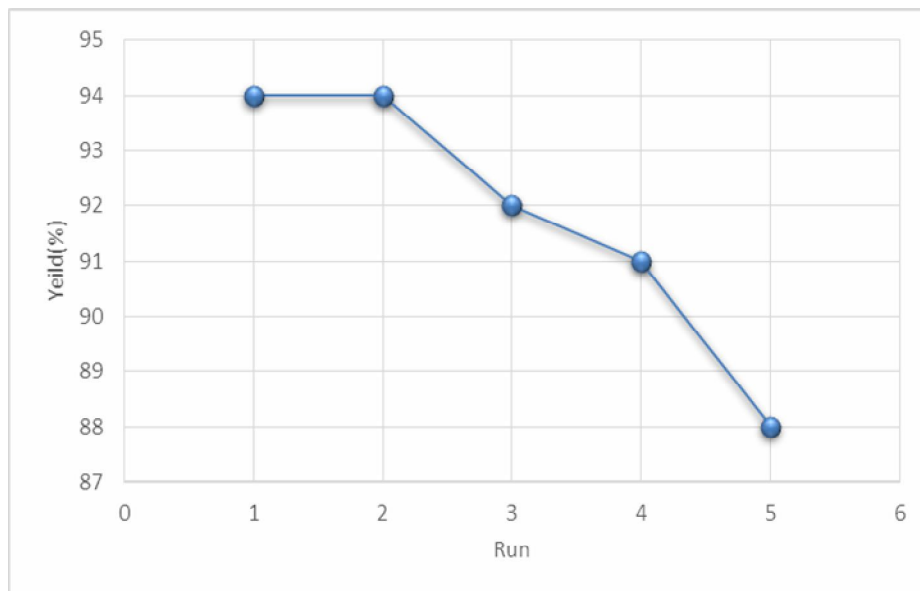


Fig. 5. Reusability of Al₂O₃ using the model reaction.

supporting this work by Grant (No. 256722/26).

REFERENCES

- [1] a) A. Ayati, S. Emami, A. Asadipour, A. Shafiee, A. Foroumadi *Eur J. Med. Chem.* 97 (2015) 699; b) E. Theophil, H. Siegfried, *The Chemistry of Heterocycles: Structure, Reactions, Syntheses and Applications* ISBN: 3-527-30720-6; c) A. Kleemann, *J. Engel. Pharmaceutical Substances*, 4th Ed., 2001.
- [2] M.T. Frija, A.J.L. Pombeiro, M.N. Kopylovich, *Coord. Chem. Rev.* 308 (2016) 32.
- [3] M.A. Khanfar, D. Reiner, S. Hagenow, H. Stark, *Bioorg. Med. Chem.* 26 (2018) 4034.
- [4] J. Safari, Z. Abedi-Jazini, Z. Zarnegar, M. Sadeghi, *J. Nanopart. Res.* 17 (2015) 495.
- [5] J.V. Metzger, *Thiazole and its Derivatives*, Wiley, New York, 1979.
- [6] T. Iino, D. Tsukahara, K. Kamata, K. Sasaki, S. Ohyama, H. Hosaka, T. Hasegawa, M. Chiba, Y. Nagata, J. Eiki, T. Nishimura, *Bioorg. Med. Chem.* 17 (2009) 2733.
- [7] J. Kim, Y. Moon, S.W. Ham, *Korean. Chem. Soc.* 32 (2011) 2893.
- [8] T.K. Venkatachalam, E.A. Sudbeck, C. Mao, F.M. Uckun, *Bio. Med. Chem.* 11 (2001) 523.
- [9] B.F. Abdel-Wahab, S.F. Mohamed, A.E.G.E. Amr, M.M. Abdalla, *Monatsh. Chem.* 139 (2008) 1083.
- [10] A.D. Logu, M. Saggi, M.C. Cardia, R. Borgna, C. Sanna, B. Saggi, E.J. Maccioni, *Antimicro. Chem.* 55 (2005) 692.
- [11] N. Singh, S.K. Bhati, A. Kumar, *Eur. J. Med. Chem.* 43 (2008) 2597.
- [12] D. Bhuniya, R. Mukhhavilli, R. Dhivhare, D. Launay, R. Dere, A. Deshpandey, A. Verma, P. Vishwakarma, M. Moger, A. Pradhan, H. Pati, V.S. Gopinath, S. Gupta, S. Puri, D. Martin, *Eur. J. Med. Chem.* 102 (2015) 582.
- [13] N. Siddiqui, W. Ahsan, *Eur. J. Med. Chem.* 45 (2010) 1536.
- [14] D. Das, P. Sikdar, M. Bairagi, *Eur. J. Med. Chem.* 109 (2016) 89.
- [15] J. Zhao, J. Xu, J. Chen, M. He, X. Wang, *Tetrahedron* 71 (2015) 539.
- [16] Z. Abedi-Jazini, J. Safari, Z. Zarnegar, M. Sadeghi, *Polycycl. Aromat. Comp.* 38 (2018) 231.
- [17] W.J. Xue, K.L. Zheng, H.Z. Li, F.F. Gao, A.X. Wu, *Tetrahedron Lett.* 55 (2014) 4212.
- [18] J. Safari, M. Sadeghi, *Res. Chem. Intermed.* 42 (2016) 8175.

- [19] S.M. Ghodse, V.N. Telvekar, *Tetrahedron Lett.* 56 (2015) 472.
- [20] J. Noei, A.R. Khosropour, *Ultrason. Sonochem.* 16 (2009) 711.
- [21] J. Safari, Z. Shokrani, Z. Zarnegar, *Polycycl. Aromat. Comp.* In Press (2018).
- [22] H. Karade, M. Sathe, M.P. Kaushik, *Catal. Commun.* 8 (2007) 741.
- [23] T.M. Potewar, S.A. Ingale, K.V. Srinivasan, *Tetrahedron* 63 (2007) 11066.
- [24] F.M. Pedro, S. Hirner, F.E. Kühn, *Tetrahedron Lett.* 46 (2005) 7777.
- [25] A. Akbari, M. Amini, A. Tarassoli, B. Eftekhari-Sis, N. Ghasemian, E. Jabbari, *Nano-Struct. Nano-Object.* 14 (2018) 19.
- [26] J. Zhang, J.W. Medlin, *Surf. Sci. Rep.* 73 (2018) 117.
- [27] a) A. Maleki, R. Firouzi-Haji, P. Farahani, *Org. Chem. Res.* 4 (2018) 86; b) A. Maleki, A.A. Jafari, S. Yousefi, *J. Iran. Chem. Soc.* 14 (2017) 1801; c) A. Maleki, M. Aghaei, R. Paydar, *J. Iran. Chem. Soc.* 14 (2017) 485.
- [28] B.C. Gates, *Chem. Rev.* 9 (1995) 511.
- [29] A. Krell, H.W. Ma, *J. Am. Ceram. Soc.* 86 (2003) 241.
- [30] J.M. Schneider, W.D. Sproul, A.A. Voevodin, A. Matthews, *J. Vac. Sci. Technol. A. Vac. Surf. Films.* 15 (1997) 1084.
- [31] M. Trueba, S.P. Trasatti, *Eur. J. Inorg. Cheminform.* 36 (2005) 3393.
- [32] E.U. Ezugwu, J. Bonney, da Silva, *Alloy. J. Braz. Soc. Mech. Sci. Eng.* XXVI (1) (2004) 12.
- [33] W.H. Gitzen (Ed.), *Alumina as a Ceramic Material.* American Ceramic Society, Columbus, 1970.
- [34] K.Y. Paranjpe, *Pharma Innovation.* 6 (2017) 236.
- [35] A. Amoozadeh, M. Ahmadzadeh, E. Kolvari, *J. Chem.* (2013) Article ID 767825.
- [36] O. Rahmanpour, A. Shariati, M.R. Khosravi Nikou, M. Rohani, *Synth. React. Inorg. Met.-Org. Chem.* 46 (2016) 171.
- [37] M.H. Parvin, J. Arjomandi, J.Y. Lee, *Catal. Commun.* 110 (2018) 59.
- [38] A. Rajaeiyan, M.M. Bagheri-Mohagheghi, *Adv. Manuf. I* (2013) 176.
- [39] J. Li, P. Yubai, C. Xiang, Q. Ge, J. Guo, *Ceram. Int.* 32 (2006) 578.
- [40] J. Safari, Z. Abedi-Jazini, Z. Zarnegar, M. Sadeghi, *Catal. Commun.* 77 (2016) 108.
- [41] D. Caceres-Castillo, R.M. Carballo, J.A. Tzecz-Interian, G.J. Mena-Rejon, *Tetrahedron Lett.* 53 (2012) 3934.
- [42] Z. Zarnegar, R. Alizadeh, M. Ahmadzade, J. Safar, *J. Mol. Struct.* 1144 (2017) 58.
- [43] D.V. Jawale, D.L. Lingampalle, U.R. Pratap, R.A. Mane, *Chin. Chem. Lett.* 21 (2010) 412.