Res Chem Intermed (2017) 43:5985–5994
DOI 10.1007/s11164-017-2974-8

A one-pot multi-component reaction for the facile synthesis of some novel 2-aryl thiazolidinones bearing benzimidazole moiety using $La(NO_3)_3{\cdot}6H_2O$ as an efficient catalyst

Mehdi Kalhor[1] · Soodabeh Banibairami[1] · Seyed Ahmad Mirshokraie[1]

Received: 14 February 2017 / Accepted: 2 May 2017 / Published online: 18 May 2017

Abstract A series of new derivatives of 3-benzimidazolyl-2-aryl thiazolidinones, **4a–j** are synthesized via a rapid, one-pot, three-component reaction by using $La(NO_3)_3{\cdot}6H_2O$ as an efficient catalyst from the reaction of 2-aminobenzimidazole, aromatic aldehydes and thioglycolic acid in ethanol at room temperature. These new compounds were characterized by IR, ^1H, ^{13}C NMR and mass spectroscopies. An inexpensive and available catalyst, short reaction time, easy workup, good to excellent yields and nontoxic solvent are the advantages of this reaction.

Keywords Synthesis · Multi-component reaction · Catalyst · Lanthanum nitrate hexahydrate · 1,3-thiazolidinone · 2-aminobenzimidazole

Introduction

Synthesis of complicated molecules by one-pot multi-component reactions (MCRs) can be achieved in a very fast, efficient and high yield without isolation of intermediate. MCRs are convergent reactions, in which three or more starting materials react to form a product, where basically all or most of the atoms contribute to the newly formed product. In the MCRs, a product is assembled according to a cascade of elementary chemical reactions. The result is clearly dependent on the reaction conditions: solvent, temperature, catalyst, concentration, the kind of starting materials and functional groups. In general, using the catalyst is the most popular approach for the synthesis by the MCRs method for industrial applications [1–3].

1,3-Thiazolidin-4-ones having sulfur and nitrogen in a five-membered ring belong to thiazolidines which have efficient activities in biological, pharmaceutical

✉ Mehdi Kalhor
mekalhor@gmail.com; mekalhor@pnu.ac.ir

[1] Department of Chemistry, Payame Noor University, P.O. BOX 19395-4697, Tehran, Iran

and agricultural fields. They have been employed in the preparation of different important drugs such as famotidine, pramipexole and riluzole. [4, 5]. Nowadays, their antifungal, antimicrobial, anticancer, antidiabetic, antiviral and antiHIV activities have been reported [6, 7]. The popular method for the preparation of the 1,3-thizolidinone involves the condensation of an amine, an aldehyde and thioglycolic acid in one or two steps by refluxing in a toxic solvent at high temperature following by azeotropic distillation or under microwave irradiation by the drastic condition of removing the water [8–10]. Also, another way for approaching 1,3-thiazolidinone consisting of the condensation of amine, chloroacetylchloride and thiocyanate by refluxing in two steps has been reported [11]. According to the importance of this compound, several catalytic syntheses of thiazolidinones have been recently reported. These involve a coupling reagent like DCC [12], $Bi(SCH_2CO_2H)_3$, solvent-free, 80 °C [13], silica gel [14], Schiff base MCM-41/$CuSO_4$ [15], DIPEA reagent [16], Pd NPs, solvent-free [17], free catalyst in water [18], etc. [19–21]. However, most of them have limitations including long reaction times in multi-step synthesis, the use of expensive and toxic catalysts or reagents, low yields and tedious separation of the pure product from the reaction mixture. Therefore, the pursuance of more convenient and practical synthetic methods for the preparation of these compounds is still demanded as an active research area. On the other hand, it is known that, if an active core ring is linked to another nucleus, the resulting molecule may possess more powerful potential for biological activity. Also, reports show that the benzimidazole derivatives are used for their antiviral [22, 23], anti-ulcer [24], antimicrobial [25–28], antifungal [29, 30], anti-inflammatory [31], anthelmintic [32, 33], anticancer, anti-HIV-1 and antihistaminic properties [34–37].

Looking at the above facts and as a part of our ongoing studies on the catalytic synthesis of compounds containing the benzimidazole nucleus [11, 34, 38], this paper aims to report the one-pot, three-component synthesis of 2-((1*H*-benzo[*d*]imidazol-2-ylamino)(aryl) 1,3-thiazolidin-4-ones **4a–j** using $La(NO_3)_3{\cdot}6H_2O$ as an efficient catalyst with good to excellent yield in EtOH at room temperature.

Experimental

Materials and instruments

All the compounds were identified by their physical and spectroscopic data. Melting points were measured by using capillary tubes on an electrothermal digital apparatus and are uncorrected. IR spectra were recorded using a KBr disc on a galaxy series FT-IR 5000 spectrometer. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Brucker spectrophotometer (300 MHz) in DMSO-d_6 using Me_4Si as an internal standard. The mass spectra were recorded on an Agilent model: 5975C VL MSD with a Triple-Axis detector spectrometer at 70 eV. Reactions were monitored by thin layer chromatography (TLC) (Table 1).

Table 1 Optimizing the model reaction conditions of 2-aminobenzimidazole, 4-nitrobenzaldehyde and thioglycolic acid using different kinds and amounts of catalyst and solvents at room temperature

Entry	Catalyst	Catalyst loading (mol%)	Solvent	Time (min)	Yield (%)[a]
1	$La(NO_3)_3{\cdot}6H_2O$	5	EtOH	30	80
2	$La(NO_3)_3{\cdot}6H_2O$	10	EtOH	25	90
3	$La(NO_3)_3{\cdot}6H_2O$	15	EtOH	30	78
4	CAN	10	EtOH	35	33
5	PTSA	10	EtOH	35	28
6	$Ni(NO_3)_3{\cdot}6H_2O$	10	EtOH	45	85
7	$La(NO_3)_3{\cdot}6H_2O$	10	MeOH	35	78
8	$La(NO_3)_3{\cdot}6H_2O$	10	CH_3Cl	60	48
9	$La(NO_3)_3{\cdot}6H_2O$	10	MeCN	60	52
10	$La(NO_3)_3{\cdot}6H_2O$	10	Acetone	35	82
11	–	–	EtOH	60	Trace

[a] Isolated yields

General procedure for the preparation of 1,3-thiazolidinones 4a–j

To a mixture of aromatic aldehyde (1 mmol), 2-aminobenzimidazole (1.1 mmol) and thioglicolic acid (1.2 mmol) in 5 ml ethanol, $La(NO_3)_3{\cdot}6H_2O$ (10 mol%) as catalyst was added. The reaction mixture was stirred magnetically at room temperature for the appropriate time as shown in Table 2 (the progress of reaction being monitored by TLC and using *n*-hexane/ethyl acetate (2:1 v/v) as an eluent). After completion of the reaction, 5 mL water were added and the precipitated product was filtered and washed with cold ethanol–water. The residue was recrystallized from the ethanol–water and air-dried.

Spectroscopic data for new compounds

3-(1H-benzo[d]imidazol-2-yl)-2-phenylthiazolidin-4-one ***(4a)*** IR (KBr) (ν_{max}): 3360 (NH), 1700 (C=O), 1532 (C=N), 1381, 1269 (C=C), 1117 (C-N), 655 (C-S-C) cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.93 (1H, d, $J = 16.53$ Hz, SCH_2), 4.17 (1H, d, $J = 16.56$ Hz, SCH_2), 6.77 (1H, s, CH), 7.06–7.11 (2H, m, H-Ar), 7.24–7.39 (6H, m, H-Ar), 7.50 (1H, d, $J = 7.50$ Hz, H-Ar), 12.44 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.0, 61.5, 111.2, 121.6, 125.3, 127.9, 128.6, 134.4, 141.4, 144.4, 171.7 ppm; MS (m/z, %): 295.1 (M^+, 40), 249.1 (31), 220.1 (44), 133.1 (100), 105.1 (69), 77.1 (40).

3-(1H-benzo[d]imidazol-2-yl)-2-(2-hydroxyphenyl)thiazolidin-4-one ***(4b)*** IR (KBr) (ν_{max}): 3553 (OH), 3329 (NH), 1705 (C=O), 1599 (C = N), 1456, 1363, 1274 (C=C), 1231 (C-N), 669 (C-S-C) cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.88 (1H, d, $J = 16.38$ Hz, SCH_2), 4.04 (1H, d, $J = 16.44$ Hz, SCH_2), 6.68 (1H, t, $J = 7.47$ Hz, H-Ar), 6.76 (1H, s, *CH*), 6.83 (1H, d, $J = 8.01$ Hz, H-Ar), 6.92 (1H,

Table 2 Synthesis of 2-((1*H*-benzo[*d*]imidazol-2-ylamino)(Aryl) 1,3-thiazolidin-4-ones, **4a–j** in the presence of 10 mol% $La(NO_3)_3{\cdot}6H_2O$ in ethanol at room temperature

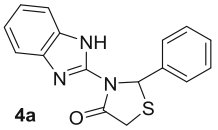
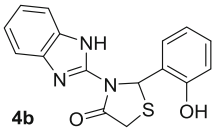
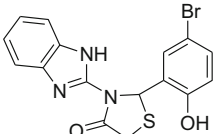
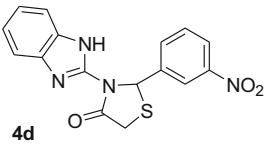
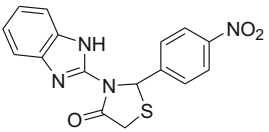
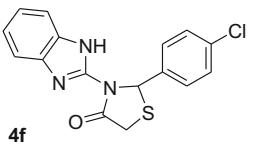
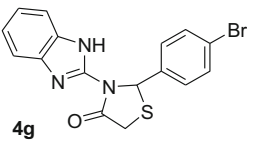
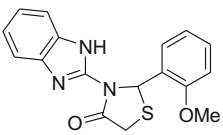
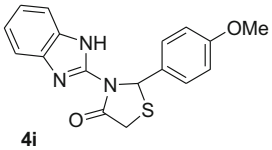
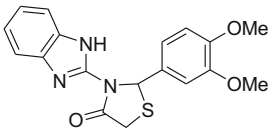
Entry	R	Product	Time (min)	M.p. (°C)	Yield (%)[a]
1	H	 **4a**	30	208-210	80
2	2-OH	 **4b**	25	245-246	85
3	2-OH-5-Br	 **4c**	30	260-261	88
4	3-NO_2	 **4d**	25	146-148	88
5	4-NO_2	 **4e**	25	207-210	90
6	4-Cl	 **4f**	25	229-230	87
7	4-Br	 **4g**	25	244-246	89
8	2-OMe	 **4h**	30	263-266	77

Table 2 continued

Entry	R	Product	Time (min)	M.p. (°C)	Yield (%)[a]
9	4-OMe	 **4i**	30	215-217	79
10	3,4-diOMe	 **4j**	30	167-170	81

[a] Isolated yields

d, $J = 7.59$ Hz, H-Ar), 7.06–7.13 (3H, m, H-Ar), 7.37 (1H, d, $J = 7.32$ Hz, H-Ar), 7.51 (1H, d, $J = 7.23$ Hz, H-Ar), 10.11 (1H, s, OH), 12.43 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.1, 57.8, 111.8, 115.6, 117.5, 118.8, 121.5, 121.6, 124.4, 127.0, 128.8, 132.7, 139.8, 144.5, 154.1, 172.1 ppm; MS (m/z, %): 311.1 (M^+, 63), 238.1 (81), 220.1 (66), 160.1 (38), 118.1 (44), 77.1 (14), 58.1 (62), 43.1 (100).

3-(1H-benzo[d]imidazol-2-yl)-2-(5-bromo-2-hydroxyphenyl)thiazolidin-4-one ***(4c)*** IR (KBr) (ν_{max}) 3412 (OH), 3348 (NH), 1690 (C = O), 1617, 1537 (C = N), 1467, 1369, 1274 (C = C), 1228 (C-N), 658 (C-S-C) cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H 3.88 (1H, d, $J = 16.38$ Hz, SCH_2), 4.13 (1H, d, $J = 16.35$ Hz, SCH_2), 6.70 (1H, s, CH), 6.78 (1H, d, $J = 8.61$ Hz, H-Ar), 7.10 (3H, d br, H-Ar), 7.23 (1H, dd, $J = 2.37$ Hz, H-Ar), 7.39–7.51 (2H, t br, H-Ar), 10.45 (1H, s, OH), 12.43 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C 32.3, 57.5, 109.9 (2C), 111.9, 117.8, 121.6, 127.3, 129.7, 131.4, 144.4, 153.6, 172 ppm; MS (m/z, %) 391.1 (M^+, 52), 316.1 (87), 216.9 (12), 160.1 (100), 118.1 (51), 91.1 (17).

3-(1H-benzo[d]imidazol-2-yl)-2-(3-nitrophenyl)thiazolidin-4-one ***(4d)*** IR (KBr) (ν_{max}): 3336 (NH), 1704 (C=O), 1620 (C=N), 1520, 1349 (NO_2), 1262 (C=C), 1119 (C-N), 617 (C-S-C) cm^{-1};^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.94 (1H, d, $J = 16.56$ Hz, SCH_2), 4.24 (1H, d, $J = 16.53$ Hz, SCH_2), 6.93 (1H, s, CH), 7.03-7.13 (2H, m, H-Ar), 7.34 (1H, d, $J = 7.50$ Hz, H-Ar), 7.50 (1H, d, $J = 7.35$ Hz, H-Ar), 7.62 (1H, t, $J = 7.98$ Hz, H-Ar), 7.85 (1H, d, $J = 7.92$ Hz, H-Ar), 8.09–8.12 (1H, d br, H-Ar), 8.29 (1H, t br, H-Ar), 12.47 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.0, 60.6, 111.9, 117.6, 120.5, 121.6, 121.8, 122.9, 130.3, 131.8, 132.7, 139.8, 143.9, 144.3, 147.9, 171.6 ppm; MS (m/z, %): 340 (M^+, 100), 294.1 (86), 265.1 (39), 219.1 (37), 164.1 (27), 118 (43), 91 (28).

3-(1H-benzo[d]imidazol-2-yl)-2-(4-nitrophenyl)thiazolidin-4-one ***(4e)*** IR (KBr) (ν_{max}): 3477 (NH), 1708 (C=O), 1617, 1514 (C=N), 1520, 1349 (NO_2), 1451, 1345, 1270 (C=C), 1114 (C-N), 626 (C-S-C) cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.96 (1H, d, $J = 16.53$ Hz, SCH_2), 4.20 (1H, d, $J = 16.53$ Hz, SCH_2), 6.90 (1H, s, CH), 7.03-7.13 (2H, m, H-Ar), 7.33 (1H, d, $J = 7.68$ Hz, H-Ar), 7.50 (1H, d, $J = 7.53$ Hz, H-Ar), 7.66 (2H, d, $J = 8.70$ Hz, H-Ar), 8.16 (2H, d, $J = 8.67$ Hz, H-Ar) 12.47 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.0, 60.7, 111.2, 121.7, 122.4, 123.2, 124.0, 126.6, 130.3, 144.3, 146.9, 149.0, 152.9, 171.5 ppm; MS (m/z, %): 340.2 (M^+, 100), 294.1 (92), 219.1 (50), 164.1 (26), 118.1 (41), 91.1 (27), 77.1 (13).

3-(1H-benzo[d]imidazol-2-yl)-2-(4-chlorophenyl)thiazolidin-4-one ***(4f)*** IR (KBr) (ν_{max}): 3324 (NH), 1708 (C=O), 1620, 1536 (C=N), 1488, 1450, 1366, 1310, 1270 (C=C), 1227, 1213, 1117 (C-N), 1108, 1009, 828, 739, 722, 658 (C-S-C), 500 cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.92 (1H, d, $J = 16.53$ Hz, SCH_2), 4.18 (1H, d, $J = 16.50$ Hz, SCH_2), 6.76 (1H, s, CH), 7.06–7.11 (2H, m, H-Ar), 7.35-7.44(5H, q br, H-Ar), 7.49 (1H, d, $J = 7.38$ Hz, H-Ar), 12.42 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.0, 60.9, 111.2, 119.7, 121.0, 121.6, 127.4, 128.3(d), 128.6, 132.1, 132.4, 140.6, 144.3, 171.6 ppm; MS (m/z, %): 329.5 (M^+, 100), 287.1 (21), 254.1 (59), 175.1 (24), 135.1 (76), 91.1 (41), 46.1 (77).

3-(1H-benzo[d]imidazol-2-yl)-2-(4-bromophenyl)thiazolidin-4-one ***(4g)*** IR (KBr) (ν_{max}): 3413 (NH), 1706 (C=O), 1618, 1535 (C=N), 1486, 1450, 1369, 1270 (C=C), 1227, 1117 (C-N), 1006, 738, 722, 659 (C-S-C), 497 cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.92 (1H, d, $J = 16.53$ Hz, SCH_2), 4.18 (1H, d, $J = 16.53$ Hz, SCH_2), 6.74 (1H, s, CH), 7.04–7.13 (2H, m, H-Ar), 7.34 (3H, d, $J = 8.37$ Hz, H-Ar), 7.49 (3H, d, $J = 8.40$ Hz, H-Ar), 12.42 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.0, 61.0, 112.0, 119.8, 121.0, 121.6, 127.6, 128.9, 131.2, 131.5, 141.0, 144.3, 171.6 ppm; MS (m/z, %): 375.1 (M^+, 25), 327.1 (23), 300.1 (100), 144.1 (10), 118.1 (35), 91.1 (28).

3-(1H-benzo[d]imidazol-2-yl)-2-(2-methoxyphenyl)thiazolidin-4-one ***(4 h)*** IR (KBr) (ν_{max}): 3378 (NH), 2961 (C-H), 1696 (C=O), 1622, 1545, 1493 (C=N), 1450, 1373 (C=C), 1270, 1251, (C-O), 1100 (C-N), 1118,753, 746, 644 (C-S-C) cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.88 (3H, s, OCH_3), 3.88 (1H, d, $J = 16.23$ Hz, SCH_2), 4.01 (1H, d, $J = 16.47$ Hz, SCH_2), 6.77 (1H, s, CH), 6.80 (1H, t, $J = 7.47$, H-Ar), 6.97 (1H, d, $J = 7.14$ Hz, H-Ar), 7.03–7.13 (3H, m, H-Ar), 7.25 (1H, t, $J = 7.47$, H-Ar), 7.36 (1H, d, $J = 7.50$, H-Ar), 7.51 (1H, d, $J = 7.41$, H-Ar), 12.44 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.0, 55.7, 57.6, 111.4, 111.9, 117.6, 120.3, 121.5, 121.7, 124.1, 128.5, 129.2, 132.8, 139.9, 144.5, 155.9, 172.0 ppm; MS (m/z, %): 326.2 (M^+, 40), 294 (100), 220.1 (40), 91.2 (19), 46.1 (15).

3-(1H-benzo[d]imidazol-2-yl)-2-(4-methoxyphenyl)thiazolidin-4-one ***(4i)*** IR (KBr) (ν_{max}): 3478 (NH), 1707 (C=O), 1638, 1617, 1540, 1511 (C=N), 1452, 1373 (C=C), 1270, 1254 (C-O), 1178 (C-N), 1116, 1026, 843, 726, 665 (C-S-C), 607 cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.68 (3H, s, OCH_3), 3.91 (1H, d,

$J = 16.53$ Hz, SCH_2), 4.17 (1H, d, $J = 16.53$ Hz, SCH_2), 6.71 (1H, s, CH), 6.83 (2H, d, $J = 8.61$ Hz, H-Ar), 7.04–7.12 (2H, m, H-Ar), 7.31 (2H, d, $J = 8.61$ Hz, H-Ar), 7.37 (1H, d, $J = 7.53$ Hz, H-Ar), 7.48 (1H, d, $J = 7.14$ Hz, H-Ar), 12.41 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.2, 55.0, 61.5, 112.0, 113.7, 113.9, 121.6, 126.9, 133.2, 144.4, 158.9, 171.6 ppm; MS (m/z, %): 325.2 (M^+, 100), 279.1 (59), 250.1 (72), 165.1 (55), 135.1 (51), 118.1 (33), 91 (22).

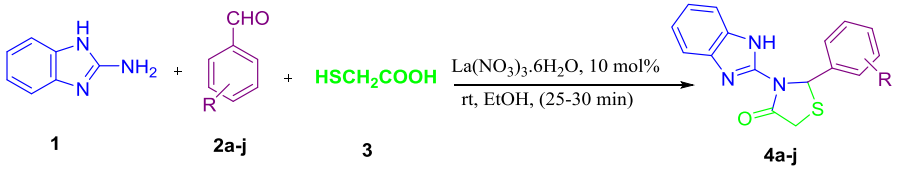
2-2-10 3-(1H-benzo[d]imidazol-2-yl)-2-(3,4-dimethoxyphenyl)thiazolidin-4-one ***(4j)*** IR (KBr) (ν_{max}): 3400 (NH), 1704 (C=O), 1637, 1617, 1537 (C=N), 1514, 1452 (C=C), 1273, 1256, 1239 (C-O), 1141 (C-N), 648 (C-S-C), 616, 478 cm^{-1}; ^{1}H-NMR (300 MHz, DMSO-d_6) δ_H: 3.67 (3H, s, OCH_3), 3.72 (3H, s, OCH_3), 3.90 (1H, d, $J = 16.50$ Hz, SCH_2), 4.16 (1H, d, $J = 16.53$ Hz, SCH_2), 6.70 (1H, s, CH), 6.81 (2H, s, H-Ar), 7.05–7.13 (3H, m, H-Ar), 7.38 (1H, d, $J = 7.47$ Hz, H-Ar), 7.48 (1H, d, $J = 7.29$ Hz, H-Ar), 12.42 (1H, s, NH) ppm; ^{13}C-NMR (75 MHz, DMSO-d_6) δ_C: 32.1, 55.4 (2C), 61.6, 109.7, 111.4, 111.8, 117.0, 117.7, 121.5, 121.7, 132.7, 133.4, 140.0, 144.4, 148.5, 148.8, 171.7 ppm; MS (m/z, %): 355.2 (M^+, 100), 313.2 (61), 280.2 (50), 192.1 (17), 165.1 (81), 118.1 (30), 91.1 (18).

Results and discussion

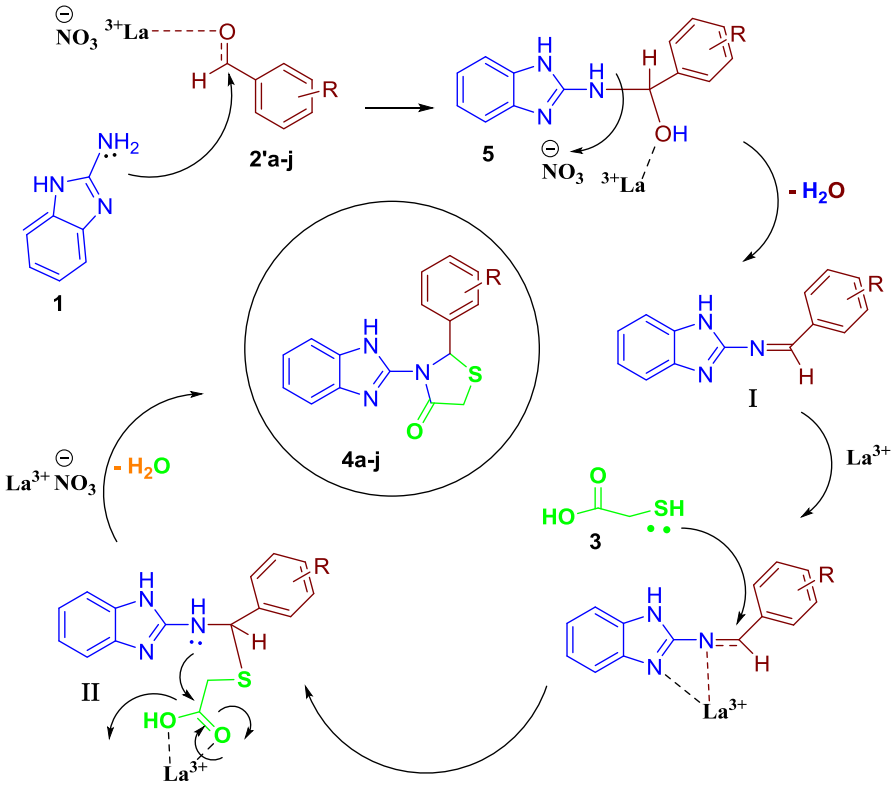
Firstly, to obtain the optimal method conditions, the effect of the solvent and the amount and type of efficient catalyst on the yield of the reaction were examined. From the data summarized in Table 1, 10 mol% of $La(NO_3)_3{\cdot}6H_2O$ was the best catalyst in ethanol resulting in the highest yield in a model reaction of 2-aminobenzimidazole, 4-nitrobenzaldehyde and thioglycolic acid at ambient temperature (Table 1, entry **2**). Also, the reaction product was evaluated in the absence of catalyst and its yield was negligible (Table 1, entry **11**).

By the generality of this method, some novel 2-((1*H*-benzo[*d*]imidazol-2-ylamino)(aryl) 1,3-thiazolidin-4-ones were prepared via one-pot multi-component reaction by employing various aromatic aldehydes under the same conditions (Scheme 1). The results are presented in Table 2.

Based on our previous knowledge, a probable reaction mechanism for the synthesis of the 2-((1*H*-benzo[*d*] imidazol-2-ylamino) (Aryl) 1,3-tiazolidin-4-ones (**4a–j**) is proposed in Scheme 2. Initially, $La(NO_3)_3{\cdot}6H_2O$ activates the carbonyl group of the aromatic aldehyde to afford intermediate **2′a–j**, and then 2-aminobenzimidazole as a nucleophile attack it to form the intermediate **5** which can followed by catalytic oxidation to form the intermediate **I.** The Schiff base **I** is a



Scheme 1 The synthetic pathway of compounds **4a–j**



Scheme 2 Proposed mechanism for the synthesis of compounds **4a–j**

stable colored powder with a high melting point which can be separated in a two-step reaction path. However, in the three-component reactions after the formation of the corresponding Schiff base, under catalytic activating and undergoing nucleophile attack by the third molecule, thioglicolic acid, the intermediate **II**, has instantly formed. Immediately, by the third catalytic activation of the intermediate **II** following intermolecular nucleophile attack and by the loss of the second water molecule, cyclization of 1,3-thiazolidin-4-one rings **4a–j** can be accomplished.

All products are new and their structures were assigned using the spectroscopic data. In the IR spectra of the compounds, two sharp bonds in the region between 3300–3480 and 1617–1708 cm^{-1} are attributed to vibrations of the NH and C=O groups. In the ^{1}H-NMR spectra, two diastereotopic protons that clearly appear as a doublet and doublet with $J = 16.23$–16.53 Hz around 3.88–4.24 ppm are attributed to the methylene group of the thiazolidin-4-one ring, and the singlet signal at 12.40 ppm is referred to the resonance of the NH proton of the benzimidazole ring. The other signals were observed in the expected region which are consistent with their structures. In the mass spectra, a molecular ion peak (25–100%) can be seen for all the compounds and, by considering the relative formula mass, the cyclization mechanism in the last step of the synthesis can be proved.

Conclusion

Some novel derivatives of 2-((1*H*-benzo[*d*]imidazol-2-ylamino)(aryl)1,3-thazolidine-4-one based on 2-aminobenzimidazole core were synthesized in a small amount of ethanol as a solvent via the one-pot multi-component reaction of 2-aminobenzimidazole, aromatic aldehydes and thioglicolic acid in the presence of a catalytic amount of $La(NO_3)_3{\cdot}6H_2O$ at room temperature. The attractive features of this procedure are its good conversions and easy workup, making it a useful and practical method for the synthesis of 2,3-substituted-1,3-thazolidine-4-ones. Also, considering all ofdvantages of this simple and readily available procedure, this chemical design can be consistent with the goal of green chemistry.

Acknowledgement We are grateful for the research commute of Chemistry Department of Payame Noor University for providing financial and technical support of this work.

References

1. L. Weber, Drug Discov. Today **7**, 143–147 (2002)
2. J.C. Menendez, Synthesis **15**, 2624 (2006). doi:10.1055/s-2006-949153
3. M.J. Climent, A. Corma, S. Iborra, RSC Adv. **2**, 16–58 (2012)
4. S. Pola, Signif. Thiazole-based Heterocycles Bioact. Syst. (2016). doi:10.5772/62077
5. T. Kvernmo, S. Härtter, E. Burger, Clin. Ther. **28**, 1065–1078 (2006)
6. A.K. Jain, A. Vaidya, V. Ravichandran, S.K. Kashaw, R.K. Agraval, Bioorg. Med. Chem. **20**, 3378–3395 (2012). **and references cited therein**
7. R.V. Patel, S.W. Park, Res. Chem. Intermed. **41**, 5599–5609 (2015)
8. P.D. Neuenfeldt, B.B. Drawanz, G.M. Siqueira, C.R.B. Gomes, S. Wardell, A.F.C. Flores, W. Cunico, Tetrahedron Lett. **51**, 3106–3108 (2010)
9. A.N. Solankee, K.P. Patel, R.B. Patel, Adv. Appl. Sci. Res. **3**, 117–122 (2012)
10. A. Rao, A. Chimirri, S. Ferro, A.M. Monforte, P. Monforte, M. Zappalà, Arkivoc **5**, 147–155 (2004)
11. A. Mobinikhaledi, N. Forughifar, M. Kalhor, M. Mirabolfathy, J. Heterocyclic Chem. **47**, 77–80 (2010)
12. T. Srivastava, W. Haq, S.B. Katti, Tetrahedron **58**, 7619–7624 (2002). **and references cited therein**
13. N. Foroughifar, S. Ebrahimi, Chin. Chem. Lett. **24**, 389–391 (2013)
14. M.P. Thakare, P. Kumar, N. Kumar, S.K. Pandey, Tetrahedron Lett. **55**, 2463–2466 (2014)
15. H.X. Pang, Y.H. Hui, K. Fan, X.J. Xing, Y. Wu, J.H. Yang, W. Shi, Z.F. Xie, Chin. Chem. Lett. **27**, 335–339 (2016)
16. A.V. Chate, A.G. Tathe, P.J. Nagtilak, S.M. Sangle, C.H. Gill, Chin.Chem. Catal. **37**, 1997–2002 (2016)
17. R.R. Harale, P.V. Shitre, B.R. Sathe, M.S. Shingare, Res. Chem. Intermed. **42**, 6695–6703 (2016)
18. M.P. Thakare, R. Shaikh, D. Tayade, RSC Adv. **6**, 28619–28623 (2016)
19. S.M. Sadeghzadeh, M. Malekzadeh, J. Mol. Liq. **202**, 46–51 (2015)
20. A.K. Yadav, M. Kumar, T. Yadav, R. Jain, Tetrahedron Lett. **50**, 5031–5034 (2009)
21. S.A. Jadhav, M.G. Shioorkar, O.S. Chavan, D.B. Shinde, R.K. Pardeshi, Heterocyclic Lett. **5**, 375–382 (2015)
22. C. Jun, X. Jiangtao, L. Xianjin, Bioorg. Med. Chem. Lett. **15**, 267–269 (2005)
23. K.T. Ashish, M. Anil, Ind. Chem. **45**, 489–493 (2006)
24. E. Carlsson, P. Lindberg, S. Unge, Chem. Br. **38**, 42–45 (2002)
25. M.E. Hassan, M.B. Alaa-eldin, M.B. Sahas, A.A. Farahat, Ind. J. Chem. **49**, 1515–1525 (2010)
26. A.K. Gülgü, A. Nurten, Farmaco **58**, 1345–1350 (2003)
27. Y.L. Luo, K. Baathulaa, V.K. Kannekanti, C.H. Zhou, G.X. Cai, Sci. China Chem. **58**, 483–494 (2015)
28. H.Z. Zhang, J.M. Lin, S. Rashid, C.H. Zou, Sci. China Chem. **57**, 807–822 (2014)
29. W. Maxwell, G. Brody, App. Microbial **21**, 944–945 (1971)

30. A.K. Ğulğun, A. Nurten, Turk. J. Chem. **30**, 223–228 (2006)
31. E.S. Lazer, M.R. Matteo, G.J. Possanza, J. Med. Chem. **30**, 726–729 (1987)
32. L. Srikanth, N. Raghunandan, R. Sambasiva, Der Pharma. Chemic. **3**, 344–352 (2011)
33. K. Sreena, R. Ratheesh, M. Rachana, M. Poornima, C. Shyni, Hygei **1**, 21–22 (2009)
34. A. Mobinikhaledi, N. Forughifar, M. Kalhor, Syn. React. Inorg. Met-org. Chem. **39**, 509–511 (2009)
35. K.U. Sadek, F. Al-Qalaf, R.A. Mekheimer, M.H. Elnagdi, Arabian J. Chem. **5**, 63–66 (2010)
36. M.M. Ramla, M.A. Omar, H. Tokuda, H.I. El-Diwoni, Bioorg. Med. Chem. **15**, 64896496 (2007)
37. Z. Kazimierczuk, D. Shugar, Nucleosides Nucleotides **8**, 1379–1385 (1989)
38. M. Kalhor, Org. Chem. Res. **1**, 59–65 (2015)