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A One-Pot Synthesis of Some Novel Ethyl 2-((1*H*-Benzo[*d*]imidazol-2-ylamino)(Aryl)methylthio)acetates by Nano-CuY Zeolite as an Efficient and Eco-Friendly Nanocatalyst

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A one-pot multicomponent reaction of aryl aldehydes, 2-amino benzimidazole and ethyl 2-mercaptoacetate is described as an efficient and environmentally method for the synthesis of some novel ethyl 2-((1*H*-benzo[*d*]imidazol-2-ylamino)(Aryl)methylthio) acetates in the presence of nano-Copper Y Zeolite (NCZ) as a catalyst in ethanol at room temperature. After optimizing of reaction conditions for this reaction, the amount of catalyst obtained was 10W%. The yield of reactions was in the range of 75-89%. The synthesized compounds **4a-j** were characterized using IR, ^{1}H NMR, ^{13}C NMR spectroscopy and Elemental Analysis. The present procedure offers several advantages, such as use of available, safety and reusable of the catalyst, high yields, short reaction times, easy and quick isolation of the product. This method may provide a developed groundwork for the synthesis of new commercial fungicide Benomyl and Carbendazim derivatives. Also, this procedure may be used as a synthetic pathway for preparation of new sulfur-bearing peptide derivatives based on 2-amino benzimidazole core under mild conditions.

Keywords: One-pot reaction, 2-Amino benzimidazole, Aryl aldehyde, Catalyst, Transition nano metal Zeolite

INTRODUCTION

The one-pot reaction is known as a reaction in which three or more easily accessible compounds are combined in a single reaction vessel [1-6]. One-pot reactions increase the performance of reactants by combing several operational steps without isolation of intermediates or changing the reaction conditions. Speed, diversity, efficiency and environmental amiability are some of the major advantages of these reactions. They have emerged as valuable tools for the preparation of structurally diverse chemical libraries of drug-like heterocyclic compounds [7-9].

It is well-known that heterocycles containing a benzimidazole moiety show a large variety of biological activities including inhibitors of HIV-1 reverse transcriptase[10], anticancer [11], anti-hepatitis C virus (HCV) [12], antiviral [13], antibacterial [14] and antifungal activities [15,16]. Thus, the synthesis of these compounds using a simple and practical method is still an active research area remains.

Furthermore, applying heterogeneous solid catalysts in organic synthesis and industrial manufacture of chemicals is interesting and important [17,18]. They have many advantages such as handling, low cost, insolubility in all organic solvents, thermal stability, nontoxic and environmental safety. In recent decades, there has been special attention in the use of solid acids as catalysts for a variety of organic reactions [19-23]. Among the various solid acid catalysts, zeolites have attracted a growing interest for their suitable acidity, thermal stability and low cost. The acidity of zeolite could depend on the Bronsted and Lewis acid sites [24]. We can decrease the number of proton sites and increase the number of Lewis sites by dehydration reaction. Also, exchange of monovalent cations with polyvalent ion creates strong Bronsted centers by the hydrolysis phenomena [25]. These processes are useful for catalytic reactions such as alcohol dehydration, acylation [26], esterification [27], Oxidation [28], Desulfurization

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[29] and cyclization [30,31].

In view of these points, and in continuation of our studies on the development of new methods for the synthesis of heterocyclic compounds [31,32], benzimidazole derivatives (one-pot synthesis by homogeneous catalyst) in particular [33], we report a clean and facile synthetic procedure for preparation of ethyl 2-((1*H*-benzo[*d*]imidazol-2-ylamino)(Aryl) methylthio)acetates, using Cu-Y zeolite as an efficient nano-catalyst.

EXPERIMENTAL

General Information

^{1}H NMR and ^{13}C NMR spectra were recorded on Brucker spectrophotometer (300 and 500 MHz) in DMSO-d_6 using Me_4Si as an internal standard. IR spectra were performed on a JASCO FT-IR 4200-A spectrophotometer. Microanalyses were performed by the Elemental Analyzer (Elemental, Vario EL II). Reaction progress was routinely monitored by thin layer chromatography (TLC) using silica gel F_{254} aluminum sheets (Merck). All chemicals were used as obtained without further purification.

Preparation of Nano-CuY Zeolite Catalyst

The NaY zeolite was synthesized with molar ratio: 16 NaOH: 1 $Al(OH)_3$: 15 SiO_2: 320 H_2O in our laboratory [34] and then, 200 ml of 0.01 M solution of metal salt ($CuCl_2$.2 H_2O) was added to 2.0 g of NaY Zeolite in a 250 ml flask. The mixture was stirred for 24 h and filtered. The obtained solid was washed with water until a colorless filtrate was observed. Then, 0.2 g of Cu-Y zeolite was put under ultrasound for 1 h to give nano size [31]. The catalyst was used without further purification.

General Procedure for the Preparation of 4a-j

To a mixture of 2-amino benzimidazole (1 mmol), ethyl 2-mercaptoacetate (1.2 mmol) and corresponding aromatic aldehyde (1 mmol) in ethanol (3 ml), nano-CuY Zeolite (10%W) was added and the reaction mixture stirred at room temperature for desired time as shown in Table 2. After completion of the reaction, the used catalyst was collected by filtration and cold water was added to the filtrate to give the product. Then, the solid product was filtered and washed with cold ethanol/water to give the compounds **4a-j**. In some cases, for further purifications, the crude products were purified by recrystallization from EtOH/H_2O.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(phenyl) methylthio)acetate (4a). White solid; IR (KBr): ν_{max} = 3384, 3292 (NH), 3059, 2997 (C-H), 1703 (CO), 1627, 1568, 1458, 1315 (C=N, C=C), 1204 (OCH_2), 1026, 727, 700 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6) δ_H: 1.07 (t, 3H, J = 7.10 Hz, CH_3), 3.39 (d, 1H, J = 15.40 Hz, SCH_2), 3.54 (d, 1H, J = 15.45 Hz, SCH_2), 3.99 (dd, 2H, J = 1.70 Hz and J = 5.44 Hz, OCH_2), 6.44 (s, 1H, CH), 6.88 (m, 2H, H-Ar), 7.16-7.18 (m, 2H, H-Ar), 7.26-7.31 (m, 1H, H-Ar), 7.36 (t, J = 7.40 Hz, 2H, H-Ar), 7.55-7.84 (m, 3H, H-Ar and NH), 9.45 (s, 1H, NH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ_C: 14.2, 33.4, 61.1, 61.2, 120.0, 122.4, 127.2, 128.3, 128.8, 129.9, 133.2, 140.3, 153.9, 170.2 ppm; Anal. Calcd. for $C_{18}H_{19}N_3O_2S$: C, 63.32; H, 5.61; N, 12.31; S, 9.39%; Found: C, 63.55; H, 5.60; N, 12.33; S, 9.37%.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(2-chloro-phenyl)methylthio)acetate (4b). White solid; IR (KBr): ν_{max} = 3389, 3306 (NH), 2974 (C-H), 1712 (CO), 1629, 1562, 1462, 1402, 1311 (C=N, C=C), 1190 (OCH_2), 1101, 1028, 731 (C-Cl) cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6) δ_H: 1.06 (t, J = 6.27 Hz, 3H, CH_3), 3.48 (d, J = 15.12 Hz, 1H, SCH_2), 3.60 (d, J = 15.69 Hz, 1H, SCH_2), 3.95 (br d, 2H, OCH_2), 6.70 (s, 1H, CH), 6.90-7.78 (m, 8H, H-Ar), 7.93 (br, 1H, NH), 10.69 (s, 1H, NH) ppm, The NH protons disappeared upon D_2O addition. ^{13}C NMR (75 MHz, DMSO-d_6) δ_C: 14.2, 33.5, 58.3, 61.3, 109.7, 115.7, 119.4, 120.6, 127.9, 128.6, 130.0, 131.9, 137.9, 153.7, 169.9 ppm; Anal. Calcd. for $C_{18}H_{18}ClN_3O_2S$: C, 57.52; H, 4.83; N, 11.18; S, 8.53 %; Found: C, 57.44; H, 4.91; N, 11.12; S, 8.63%.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(3-chloro-phenyl)methylthio) acetate (4c). White solid; IR (KBr): ν_{max} = 3372, 3304 (NH), 2981, 1712 (CO), 1628, 1566, 1456, 1309 (C=N, C=C), 1192 (OCH_2), 1030, 743 (C-Cl), 708 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6) δ_H 1.07 (t, 3H, J = 7.10 Hz, CH_3), 3.42 (d, 1H, J = 15.50 Hz, SCH_2), 3.55 (d, 1H, J = 15.50 Hz, SCH_2), 3.98 (dd, 2H, J = 3.65 and J = 5.15 Hz, OCH_2), 6.42 (s, 1H, CH), 6.89-6.90 (m, 2H, H-Ar), 7.16-7.20 (m, 2H, H-Ar), 7.36-7.51 (m, 3H, H-Ar), 7.63 (s, 1H, H-Ar), 7.85 (br, 1H, NH), 10.75 (br, 1H, NH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ_C: 14.2, 33.4, 60.5, 61.3, 126.0, 127.0, 128.2, 129.1 130.7, 132.7, 133.5, 142.9, 153.8,

164.2, 170.0 ppm; Anal. Calcd. for $C_{18}H_{18}ClN_3O_2S$: C, 57.52; H, 4.83; N, 11.18; S, 8.53 %; Found: C, 58.04; H, 4.94; N, 11.09; S, 8.63 %.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(4-chlorophenyl)methylthio)acetate (4d). White solid; IR (KBr): ν_{max} = 3327, 3121 (NH), 2984 (C-H), 1724 (CO), 1629, 1577, 1461, 1404, 1300 (C=N, C=C), 1190 (OCH_2), 1014, 733 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ_H: 1.05 (br t, 3H, CH_3), 3.44 (br t, 2H, SCH_2), 3.97 (brs, 2H, OCH_2), 6.42 (s, 1H, CH), 6.89-7.56 (m, 8H, H-Ar), 8.07 (s, 1H, NH), 10.76 (br, 1H, NH) ppm; ^{13}C NMR (75 MHz, DMSO-d_6) δ_C: 14.2, 33.3, 60.4, 61.2, 119.9, 128.8, 129.7, 131.5, 132.8, 139.4, 153.8, 164.5, 170.1 ppm; Anal. Calcd. for $C_{18}H_{18}ClN_3O_2S$: C, 57.52; H, 4.83; N, 11.18; S, 8.53 %; Found: C, 57.50; H, 4.73; N, 11.26; S, 8.49%.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(4-nitrophenyl)methylthio)acetate (4e). Yellow solid; IR (KBr): ν_{max} = 3321, 3109 (NH), 3022, 2991 (C-H), 1730 (CO), 1583, 1465, 1346, 1302 (C=N, C=C), 1167 (OCH_2), 733 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ_H: 1.06 (t, J = 7.08 Hz, 3H, CH_3), 3.48 (d, J = 15.66 Hz, 1H, SCH_2), 3.61 (d, J = 15.60 Hz, 1H, SCH_2), 3.97 (q, J = 6.94 Hz, 2H, OCH_2), 6.54 (s, 1H, CH), 6.90-7.19 (m, 4H, H-Ar), 7.82 (d, J = 8.40, 2H, H-Ar), 8.02 (s, 1H, NH), 8.25 (d, J = 8.31 Hz, 2H, H-Ar), 10.84 (s, 1H, NH) ppm, The NH protons disappeared upon D_2O addition. ^{13}C NMR (75 MHz, DMSO-d_6) δ_C: 14.2, 33.3, 40.5, 61.8, 112.0, 120.9, 124.1, 124.5, 128.4, 131.0, 147.4, 153.1, 170.5 ppm; Anal. Calcd. for $C_{18}H_{18}N_4O_4S$: C, 55.95; H, 4.70; N, 14.50; S, 8.30%; Found: C, 55.84; H, 4.78; N, 14.53; S, 8.25%.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(4-bromophenyl)methylthio)acetate (4f). Yellow solid; IR (KBr): ν_{max} = 3333, 3121 (NH), 3005 (C-H), 1722 (CO), 1585, 1402, 1300, 1188 (OCH_2), 733 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ_H: 1.06 (br d, 3H, CH_3), 3.38 (brq, 2H, SCH_2), 3.85 (brd, 2H, OCH_2), 6.4 (s, 1H, CH), 6.88-7.98 (m, 7H, H-Ar and NH), 10.80 (br, 1H, NH) ppm; ^{13}C NMR (75 MHz, DMSO-d_6) δ_C: 14.2, 33.3, 60.5, 61.2, 112.8, 120.0, 121.3, 129.4, 131.7, 132.6, 139.7, 153.8, 170.1 ppm; Anal. Calcd. for $C_{18}H_{18}BrN_3O_2S$: C, 51.43; H, 4.32; N, 10.00; S, 7.63 %; Found: C, 51.40; H, 4.38; N, 9.93; S, 7.54 %.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(4-methylphenyl)methylthio)acetate (4g). Yellow solid; IR (KBr): ν_{max} = 3324, (NH), 3053, 2989 (C-H), 1725 (CO), 1628, 1601, 1574, 1524, 1464 (C=N, C=C), 1296 (OCH_2), 1174, 1040, 735 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6) δ_H: 1.06 (t, 3H, J = 7.10 Hz, CH_3), 2.41 (s, 3H, Me), 3.35 (d, 1H, J = 15.40 Hz, SCH_2), 3.50 (d, 1H, J = 15.40 Hz, SCH_2), 3.97 (q, J = 5.55 Hz, 2H, OCH_2), 6.88 (s, 1H, CH), 7.15-7.19 (m, 3H, H-Ar), 7.38-7.43 (m, 3H, H-Ar), 7.71 (br, 1H, NH), 7.94 (d, J = 8.00 Hz, 2H, H-Ar), 9.39 (s, 1H, NH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ_C: 14.3, 21.8, 33.4, 60.9, 61.2, 120.0, 122.3, 127.0, 129.3, 130.0, 130.1, 133.0, 143.6, 165.6 ppm; Anal. Calcd. for $C_{19}H_{21}N_3O_2S$: C, 64.20; H, 5.96; N, 11.82; S, 9.02 %; Found: C, 63.45; H, 5.98; N, 11.79; S, 9.03 %.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(4-methoxyphenyl)methylthio) acetate (4h). Yellow solid; IR (KBr): ν_{max} = 3335, 3119 (NH), 3065, 2980 (C-H), 1723 (CO), 1599, 1578, 1526, 1464, 1381 (C=N, C=C), 1297, 1250, 1185 (OCH_2, C-O), 1032, 836, 745 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6) δ_H: 1.20 (t, 3H, J = 7.00 Hz, CH_3), 3.71 (d, 1H, J = 15.10 Hz, SCH_2), 3.87 (s, 4H, SCH_2, OMe), 4.10 (dd, J = 7.05 Hz, J = 7.65 Hz, 2H, OCH_2), 6.80 (brs, 1H, CH), 7.11-7.15 (m, 4H, H-Ar), 7.40-7.54 (m, 2H, H-Ar), 8.01 (d, J = 8.10, 2H, H-Ar), 9.36 (s, 1H, NH), 10.53 (br, 1H, NH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ_C: 14.5, 38.4, 56.0, 60.9, 95.9, 115.1, 118.8, 119.1, 122.0, 122.2, 128.4, 131.9, 163.5, 164.9 ppm; Anal. Calcd. for $C_{19}H_{21}N_3O_3S$: C, 61.44; H, 5.70; N, 11.31; S, 8.63%; Found: C, 61.61; H, 5.73; N, 11.34; S, 8.56%.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino)(2-hydroxyphenyl)methylthio)acetate (4i) Yellow solid; IR (KBr): ν_{max} = 3391, 3291 (NH, OH), 3016, 2970 (C-H), 1705 (CO), 1587, 1535, 1533, 1465, 1384 (C=N, C=C), 1194 (OCH_2), 1109, 727 cm^{-1}; ^1H NMR (300 MHz, DMSO-d_6) δ_H: 1.04 (t, J = 6.84 Hz, 3H, CH_3), 3.42 (d, J = 15.12 Hz, 1H, SCH_2), 3.55 (d, J = 15.24 Hz, 1H, SCH_2), 3.95 (dd, 2H, OCH_2), 6.62 (s, 1H, CH), 6.80-7.58 (m, 10H, H-Ar, NH and OH), 10.59 (br, 1H, NH) ppm, The NH and OH protons disappeared upon D_2O addition; ^{13}C NMR (75 MHz, DMSO-d_6) δ_C: 14.2, 33.7, 56.8, 61.1, 116.4, 119.4, 120.1, 126.7, 127.9, 129.4, 132.8, 154.4, 161.0, 165.9, 170.3 ppm; Anal. Calcd. for $C_{18}H_{19}N_3O_3S$: C, 60.49; H, 5.36; N, 11.76; S, 8.97%; Found: C, 60.58; H, 5.42; N, 11.64; S, 8.95%.

Ethyl 2-((1H-benzo[d]imidazol-2-ylamino) (5-bromo-2-hydroxyphenyl)methylthio) acetate (4j). Yellow solid; IR (KBr): ν_{max} = 3385, 3296 (NH, OH), 3016, 2980 (C-H),

1708 (CO), 1601, 1569, 1462, 1409, 1374, 1311 (C=N, C=C), 1282, 1243, 1191 (C-O, OCH_2), 1092, 1020, 816 (C-Br), 742 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6) δ_H: 1.07 (t, J = 7.10 Hz, 3H, CH_3), 3.42 (d, J = 15.23 Hz, 1H, SCH_2), 3.54 (d, J = 15.35 Hz, 1H, SCH_2), 4.00 (m, 2H, OCH_2), 6.80 (s, 1H, CH), 6.89-6.92 (m, 2H, H-Ar), 7.16-7.20 (m, 3H, H-Ar), 7.54-7.69 (m, 4H, H-Ar, NH and OH), 10.71 (br, 1H, NH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6) δ_C: 14.2, 33.6, 56.0, 61.1, 110.5, 118.5, 119.7, 120.1, 127.1, 128.3, 128.8, 129.4, 131.8, 140.2, 153.8, 170.3 ppm; Anal. Calcd. for $C_{18}H_{18}BrN_3O_3S$: C, 49.55; H, 4.16; Br, 18.31; N, 9.63; S, 7.35%; Found: C, 49.30; H, 4.14; N, 9.60; S, 7.37%.

RESULTS AND DISCUSSION

First, nano-Copper Y-Zeolite (NCZ) was prepared in our laboratory by the use of previously reported ultrasound-assisted procedure (31,34). The CuY zeolites were initially produced under ultrasound to obtain nano-size. The scanning electron microscopy (SEM) image of the CuY zeolite nano-particles is shown in (Fig. 1). The particle size was mainly about 139 nm.

Then, to evaluate the synthetic potential of the proposed procedure and to optimize the amounts of catalyst and reaction conditions, the reaction of 2-amino benzimidazole, 4-nitrobenzaldehyde and ethyl 2-mercaptoacetate was examined as a model in different solvents and in the presence of various W% of NCZ as a catalyst at room temperature (Table 1).

As can be seen from Table 1, the best result was obtained when the reaction was carried out in the presence of 10W% of NCZ in ethanol as a solvent (Table 1, Entry 3). Encouraged by the above results and the development and generality of this simple method, we synthesized several ethyl 2-((1*H*-benzo[*d*]imidazol-2-ylamino)(Aryl) methyl-thio)acetates **4a-j**. This reaction was carried out with employing of 2-amino benzimidazole (**1**), ethyl 2-mercaptoacetate (**3**) and corresponding aromatic aldehyde bearing electron-withdrawing groups or electron-donating groups **2a-j** in ethanol at room temperature (Scheme 1).

The results are summarized in Table 2. The yields of reactions using this practical procedure for preparation of products **4a-j** are quite fair, the workup is very simple, amount of consumer catalyst is low, and reaction times are short. On the other hand, the reaction of 4-nitrobenzaldehyde, 2-amino benzimidazole, and ethyl 2-mercaptoacetate with Cu-Y Zeolite as a catalyst was also examined under the same experimental conditions. The results obtained were compared with the ones obtained using nano Cu-Y Zeolite as a catalyst which showed the NCZ catalyst was a fairly good catalyst for this reaction.

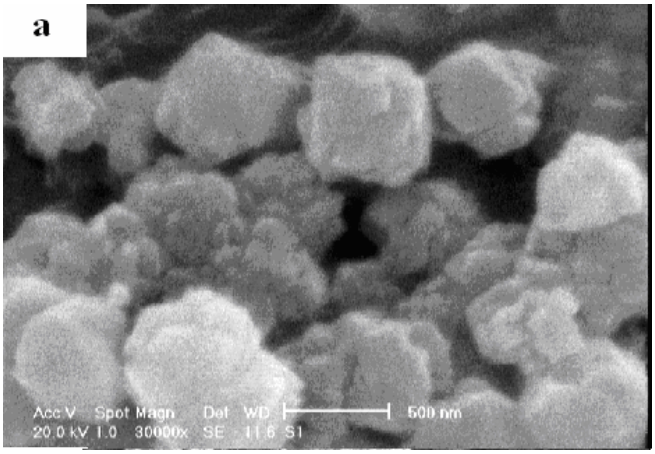


Fig. 1. The SEM photograph of the CuY zeolite nano-particles.

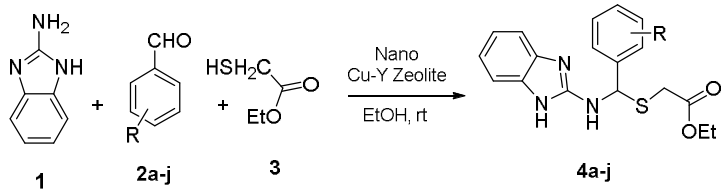
For investigation of the reusable and recycling of the catalyst, we stirred a mixture of *p*-nitrobenzaldehyde (1 mmol), 2-amino benzimidazole (1 mmol) and ethyl 2-mercaptoacetate (1.2 mmol) in the presence of nano-CuY Zeolite (10W%) at room temperature for 35 min. After completion of the reaction, the catalyst was separated through simple filtration, recovered by refluxing in ethanol for 8 h and dried at oven to 100 °C and reused in subsequent reactions with a small decrease in activity even after the fifth run (Table 3).

The structure of the products was assigned using the spectroscopic data. In the IR spectra of compounds, the two sharp bonds at the region between 3109-3391 cm^{-1} and a strong bond at the region between 1705-1730 cm^{-1} are attributed to the vibrations of two NH groups and C=O group of ester, respectively. In the ^1H NMR spectra, the singlet signal at the range of 9.39-10.84 ppm is due to the resonance of the NH proton of benzimidazole ring. The ^{13}C NMR spectra of compounds **4a-j** showed signals between 14.2-61.8 and 109.0-165.9 ppm caused by the resonance of

Table 1. Optimization of the Reaction Conditions for Preparation of Compounds **4e** Using Different Amounts of Nano-CuY Zeolite Catalyst and Different Solvents at Room Temperature

Entry	Catalyst loading (W%)	Solvent	Time (min)	Yield (%)[a]
1	2	EtOH	100	50
2	5	EtOH	85	71
3	10	EtOH	35	87
4	15	EtOH	60	61
5	10	DMF	60	72
6	10	MeCN	85	52
7	10	CH_2Cl_2	90	35
8	10	Toluene	120	15

[a]Isolated yield.



Scheme 1.The synthetic pathway of compounds **4a-j**

Table 2. Synthesis of Ethyl 2-((1*H*-benzo[*d*]imidazol-2-ylamino) (Aryl) Methylthio) Acetates, **4a-j** in the Presence of NCZ Catalyst

Entry	R	Product	Time (min)	M.P. (°C)[a]	Yield (%)[b]
1	H	**4a**	40	132	81
2	2-Cl	**4b**	35	164-166 (165)[c]	89
3	3-Cl	**4c**	25	133	75
4	4-Cl	**4d**	25	182 (184-187)	80
5	4-NO_2	**4e**	35	188-190 (196-198)	87
6	4-Br	**4f**	38	210 (216-219)	79
7	4-Me	**4g**	25	173-174	83
8	4-OMe	**4h**	30	175-177	82
9	2-OH	**4i**	40	193 (191-194)	85
10	5-Br-2-OH	**4j**	37	226	88

[a]Melting points are not corrected. [b]Isolated yield. [c]Melting points in bracketed are reported in our previous article [33].

Table 3. Catalyst Recovery Study in the Multi-component Reaction of *p*-Nitro benzaldehyde, 2-Amino Benzimidazole, and Ethyl 2-Mercaptoacetate in the Presence of Nano-CuY Zeolite (10 W%)

Entry	Time (min)	Yield (%)[a]
1	35	87
2	35	84
3	40	80
4	40	77
5	65	72

[a]Isolated yields.

aliphatic and aromatic aryl carbons. The appearance of signals at the region between 164.9-170.5 ppm attributed to the carbon resonance of the C=O group of ester is in support of the expected structures. The other signals were observed at the expected regions.

CONCLUSIONS

In conclusion, we have demonstrated a new procedure for synthesis of ethyl 2-((1*H*-benzo[*d*]imidazol-2-ylamino) (Aryl)methylthio) acetates from the multicomponent reaction of *p*-nitrobenzaldehyde, 2-amino benzimidazole, and ethyl 2-mercaptoacetate using nano-CuY Zeolite as a catalyst in ethanol at room temperature. The attractive features of this method are good conversion, reusability of catalyst, and easy work-up, making it a useful procedure for the synthesis of benzimidazole derivatives.

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