Boric acid Catalyzed Efficient Synthesis of Symmetrical *N,N'*-Alkylidene Bisamides

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Symmetrical *N,N'*-alkylidene bisamides were synthesized in excellent yield by condensation of aromatic aldehydes and amides in the presence of boric acid as a catalyst under thermal and neat microwave irradiation conditions. It is the first successful report of boric acid has been used as Lewis acid catalyst for the preparation of symmetrical *N,N'*-alkylidene bisamides. The remarkable advantages offered by this method are green catalyst, mild reaction condition, simple procedure, fast reaction and moderate to good yield of products.

Keywords: *N,N'*-Alkylidene bisamides, Amides, Aldehydes, Boric acid

INTRODUCTION

Amides and polyamides have been found as a key component of many biologically active and pharmaceutical compounds. In particular, symmetrical and unsymmetrical *N,N'*-alkylidene bisamides and their derivatives are found as key structural sub-units for the construction of peptidomimetic frameworks [1,2]. Generally symmetrical alkylidene bisamides are synthesized by the direct reaction of aldehydes with the corresponding amides and similarly unsymmetrical alkylidene bisamides are prepared from aldehydes with different amides under suitable catalytic condition. Bronsted acids such as acetic acid [3], hydrochloric acid [4] and sulfuric acid [5] have been employed to catalyze the synthetic transformation, although the results were not satisfying. Trifluromethanesulfonic acid is a more efficient catalyst in the formation of symmetrical bisamides [6]. Zhu *et al*. have reported the synthesis of fluorine-containing *N,N'*-alkylidene bisamides in the presence of fluoroalkanesulfonic acids [7].

Recently, Perumal *et al*. have reported an alternative approach to synthesis symmetrical *N,N'*-alkylidene bisamides by the reaction of aldehydes with nitriles in the presence of sulfamic acid, however the yields are moderate [8]. Milenkovic *et al*. have synthesized activated imines and aminal derivatives as potential precursors for the synthesis of β-amino acid using Dean-Stark water trap [9]. Zav'yalov *et al*. have reported the condensation of amides with carbonyl compounds in the presence of DMF-chlorotrimethylsilane as a catalyst system [10]. Bhatnagar *et al*. have reported the synthesis of benzylidene bisamides from direct condensation of benzaldehyde and different amide derivatives [11].

Synthetic methodologies based on green chemistry processes are of increasing interest in organic synthesis. In recent years, boric acid have gained special attention as a catalyst in organic synthesis for a number of synthetic transformations, for example, aza Michael addition in water [12], the thia Michael addition in water [13], Biginelli reaction [14], transesterification of ethyl acetoacetate and decarboxylation of cyclic β-enamino ketoesters [15], condensation of imidazoles, amides and aldehydes to get

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iminobenzamides are aroylated with acid chlorides to give N-[alpha-(benzylidenamino)benzyl] benzamides and *N,N'*-benzylidenebis(benzamides) [17,18]. The advantages of the boric acid catalyst are excellent solubility in water, uncomplicated handling, inexpensiveness and eco-friendly nature. Recently, several synthetically useful organic transformations using boric acid as a catalyst have also been reported in the literature [19,20].

According to the literature, a general and green chemistry method for the synthesis of symmetrical bisamides is still demanded and there is no report on the formation of symmetrical bisamides catalyzed by eco-friendly boric acid catalyst. Hence, herein we report a convenient and efficient method for the synthesis of symmetrical bisamides by condensing aryl aldehydes and aryl amides using boric acid as a catalyst.

EXPERIMENTAL

Chemicals and Apparatus

Melting points were determined in open capillaries and are uncorrected. FT-IR spectra were recorded on a Thermo Mattson Satellite FT-IR spectrophotometer by KBr pellet method. ^{1}H and ^{13}C NMR (300.1 and 75.3 MHz) spectra were recorded on a Bruker spectrometer in DMSO-d_6 as a solvent using tetramethylsilane (Me$_4$Si) as internal standard. Chemical shifts (δ) are reported in ppm. Mass spectra were recorded on Q-TOF Micromass-UK spectrometer. Chromatography purification was conducted by column chromatography using neutral alumina. Solvents used for purification are of commercial grade and purified before use [21].

General Procedure for the Preparation of *N,N'*-Alkylidene Bisamides under Conventional Heating

A mixture of aldehyde (2 mmol) and amide (4 mmol), boric acid (0.3 mmol) was added in anhydrous toluene (10 ml). The mixture was stirred at 120 °C in an oil bath and the reaction was followed by TLC analysis (eluent:n-hexane/ethyl acetate, 3:2). After completion of the reaction, the mixture was diluted with Et$_2$O (20 ml) and the catalyst allowed to settle down. The supernatant ethereal solution was decanted off, the catalyst washed with Et$_2$O (5 ml) and the combined ethereal solution concentrated under vacuum to afford the product,

which was subjected to column chromatography on neutral alumina using mixture of gradient hexane-ethyl acetate as eluent to give pure products. The symmetrical *N,N'*-alkylidene bisamides were further purified by recrystalization from ethanol.

General Procedure for the Preparation of *N,N'*-Alkylidene Bisamides under Microwave Irradiation

A mixture of aldehyde (2 mmol) and amide (4 mmol) with boric acid (0.3 mmol) in dichloromethane was made as slurry. The slurry was irradiated in an Microwave oven (SAMSUNG, 650 W) for 0-40 min at 80% Power Level (PL). The progress of the reaction was monitored by TLC analysis by taking a small amount of compound under constant time interval (eluent:n-hexane/ethyl acetate, 3:2). Further purification of the compounds has been carried out as described in the conventional heating.

Spectral Data of Compounds

N,N'-(Phenylmethylene) dibenzamide (3a). White solid, m.p.: 235 °C (lit. 237-238 °C [6]); IR (KBr): ν$_{max}$ = 3276, 1649, 1539, 1484, 1342, 1271, 1046, 706 cm^{-1}; ^{1}H NMR (300.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 7.08 (1H, t, *J* = 7.7 Hz, CH), 7.33-7.60 (11H, m, aroma), 7.93 (4H, d, *J* = 7.5 Hz, CH), 9.07 (2H, d, *J* = 7.8 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 165.6, 140.2, 133.8, 131.6, 128.3, 127.7, 127.4, 126.4, 58.6; FAB Mass calcd. for C$_{21}$H$_{18}$N$_2$O$_2$: *m/e* 353 (M + Na); found: 353.

N,N'-(4-Methylphenylmethylene) dibenzamide (3c). White solid, m.p.: 240 °C (lit. 242-243 °C [6]); IR (KBr): ν$_{max}$ = 3254, 2920, 2849, 1644, 1512, 1282, 1057, 701 cm^{-1}. ^{1}H NMR (300.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 2.29 (3H, s, CH$_3$), 7.00 (1H, t, *J* = 7.5 Hz, CH), 7.19 (2H, d, *J* = 7.8 Hz, CH), 7.35 (2H, d, *J* = 7.8 Hz, CH), 7.46-7.58 (6H, m, aroma), 7.89 (4H, d, *J* = 8.1 Hz, CH), 8.96 (2H, d, *J* = 7.5 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 165.5, 137.3, 136.8, 133.9, 131.6, 128.8, 128.3, 127.4, 126.3, 58.5, 20.6. FAB Mass calcd. for C$_{22}$H$_{20}$N$_2$O$_2$: *m/e* 367 (M + Na); found: 367.

N,N'-(4-Cyanophenylmethylene) dibenzamide (3d). White solid, m.p.: 230-234 °C; IR (KBr): ν$_{max}$ = 3276, 1654, 1556, 1480, 1331, 1282, 1073, 706 cm^{-1}. ^{1}H NMR (300.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 7.05 (1H, t, *J* = 7.5 Hz,

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CH), 7.48-7.61 (6H, m, aroma), 7.67 (2H, d, *J* = 8.1 Hz, CH), 7.88 (2H, d, *J* = 8.4 Hz, CH), 7.93 (4H, d, *J* = 7.8 Hz, CH), 9.19 (2H, d, *J* = 7.5 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 165.9, 145.5, 133.5, 132.3, 131.7, 128.3, 128.2, 127.6, 127.5, 127.4, 118.7, 110.4, 58.6. FAB Mass calcd. for C$_{22}$H$_{17}$N$_3$O$_2$: *m/e* 378 (M + Na); found: 378.

N,N'-(4-Nitrophenylmethylene) dibenzamide (3e). White solid, m.p.: 263-264 °C (lit. 265-267 °C [6]); IR (KBr): ν$_{max}$ = 3265, 2926, 1643, 1512, 1347, 1276, 1073, 712. ^{1}H NMR (300.1 MHz, DMSO-d$_6$): δ = 7.08 (1H, t, *J* = 7.5 Hz, CH), 7.47-7.60 (6H, m, aroma), 7.74 (2H, d, *J* = 8.7 Hz, CH), 7.92 (4H, d, *J* = 7.2 Hz, CH), 8.25 (2H, d, *J* = 8.7 Hz, CH), 9.23 (2H, d, *J* = 7.5 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-D$_6$, Me$_4$Si): δ (ppm): 165.9, 147.5, 147, 133.4, 131.7, 128.3, 127.9, 127.5, 123.5, 58.4. FAB Mass calcd. for C$_{21}$H$_{17}$N$_3$O$_4$: *m/e* 398 (M + Na); found: 398.

N,N'-(4-Fluorophenylmethylene) dibenzamide (3f). Yellowish solid, m.p.: 226-228 °C; IR (KBr): ν$_{max}$ = 3271, 2915, 2855, 1644, 1507, 1348, 1271, 1222, 1052, 827, 707 cm^{-1}. ^{1}H NMR (300.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 7.06 (1H, t, *J* = 7.8 Hz, CH), 7.32-7.58 (10H, m, aroma), 7.88 (2H, d, *J* = 8.1 Hz, CH), 7.91 (2H, d, 8.1 Hz, CH) 9.04 (2H, d, *J* = 7.8 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 167.9, 165.6, 140.2, 134.1, 133.7, 131.6, 131.2, 128.3, 128.2, 127.7, 127.4, 127.4, 126.4, 58.6. FAB Mass calcd. for C$_{21}$H$_{17}$FN$_2$O$_2$: *m/e* 371 (M + Na); found: 371.

N,N'-(4-Chlorophenylmethylene) dibenzamide (3g). White solid, m.p.: 230-232 °C; IR (KBr): ν$_{max}$ = 3277, 2926, 2855, 1644, 1540, 1490, 1348, 1266, 1068, 800, 701 cm^{-1}. ^{1}H NMR (300.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 7.05 (1H, t, *J* = 7.5 Hz, CH), 7.46- 7.54 (10H, m, aroma), 7.90 (4H, d, *J* = 7.5 Hz, CH), 9.09 (2H, d, *J* = 7.8 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 165.7, 139.2, 133.6, 132.3, 131.7, 128.6, 128.4, 128.3, 128.2, 127.5, 58.3. FAB Mass calcd. for C$_{21}$H$_{17}$ClN$_2$O$_2$: *m/e* 388 (M + Na); found: 388.

N,N'-(Pyridin-4-ylmethylene) dibenzamide (3h). Brown solid, m.p.: 208-212 °C; IR (KBr): ν$_{max}$ = 3277, 2926, 2855, 1647, 1540, 1460, 1320, 1276, 1075, 970, 719 cm^{-1}. ^{1}H NMR (300.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 7.05 (1H, t, *J* = 7.8 Hz, CH), 7.34-7.68 (10H, m, aroma), 7.90 (4H, d, *J* = 7.2 Hz, CH), 9.05 (2H, d, *J* = 7.8 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 165.6, 140.2, 133.7, 131.6, 128.3, 127.7, 127.4, 126.4, 58.6. FAB Mass calcd. for C$_{20}$H$_{17}$N$_3$O$_2$:

m/e 354 (M + Na); found: 354.

N,N'-(4-Methoxyphenylmethylene) dibenzamide (3j). Yellowish solid, m.p.: 230-232 °C (lit. 232-234 °C [6]) °C; IR (KBr): ν$_{max}$ = 3259, 3083, 2925, 2854, 1652, 1543, 1482, 1340, 1273, 1140, 1059, 909, 697 cm^{-1}. ^{1}H NMR (300.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 3.74 (3H, s, OCH$_3$), 6.93-7.02 (4H, m, CH), 7.38-7.56 (7H, m, aroma), 7.90 (4H, d, *J* = 8.1 Hz, CH), 8.99 (2H, d, *J* = 7.8 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 165.5, 158.8, 133.8, 132.3, 131.6, 128.3, 127.7, 127.4, 113.6, 58.3. FAB Mass calcd. for C$_{22}$H$_{20}$N$_2$O$_3$: *m/e* 383 (M + Na); found: 383.

N,N'-(Thiophen-2-ylmethylene) dibenzamide (3k). Yellowish solid, m.p.: 208-210 °C; IR (KBr): ν$_{max}$ = 3264, 3062, 2925, 2852, 1645, 1550, 1506, 1342, 1249, 1140, 1056, 931, 804, 702 cm^{-1}. ^{1}H NMR (300.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 7.00-7.035 (1H, m, CH), 7.121-7.131 (1H, m, CH), 7.27 (1H, t, *J* = 7.8 Hz, CH), 7.40-7.60 (7H, m, aroma), 7.88-7.91 (4H, m, CH), 9.19 (2H, d, *J* = 7.8 Hz, NH). ^{13}C NMR (75.3 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 170.8, 149.5, 138.8, 137.0, 133.7, 133.6, 132.7, 132.2, 131.7, 130.7, 130.1, 60.4. FAB Mass calcd. for C$_{19}$H$_{16}$N$_2$O$_2$S: *m/e* 359 (M + Na); found: 359.

N,N'-(Dimethylaminophenylmethylene) dibenzamide (3l). Brown solid, m.p.: 210-212 °C; IR (KBr): ν$_{max}$ = 3279, 2923, 1650, 1533, 1273, 1139, 1051, 805, 705 cm^{-1}. ^{1}H NMR (500.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 2.5 (6H, s, CH$_3$), 7.01 (1H, t, *J* = 8.1 Hz, CH), 7.34-7.53 (10H, m, aroma), 7.87 (4H, d, J = 6.9 Hz, CH), 8.99 (2H, d, *J* = 8.4 Hz, NH). ^{13}C NMR (125.7 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 162.3, 145.4, 131.9, 130.7, 128.9, 128.0, 127.5, 119.2, 59.5. FAB Mass calcd. for C$_{23}$H$_{23}$N$_3$O$_2$: *m/e* 396 (M + Na); found: 396.

N,N'-(3, 4-Dimethoxyphenylmethylene) dibenzamide (3m). Yellowish solid, m.p.: 216-218 °C; IR (KBr): ν$_{max}$ = 3277, 3080, 2926, 2859, 1650, 1542, 1486, 1343, 1273, 1136, 1049, 801, 707 cm^{-1}. ^{1}H NMR (500.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 3.49 (6H, s, OCH$_3$), 7.02 (1H, t, *J* = 7.8 Hz, CH), 7.27-7.57 (9H, m, aroma), 7.87 (4H, d, *J* = 9.2 Hz, CH), 9.02 (2H, d, *J* = 8.4 Hz, NH). ^{13}C NMR (125.7 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 165.9, 151.3, 141.5, 134.7, 132.2, 128.9, 128.0, 126.9, 100.1, 58.7. FAB Mass calcd. for C$_{23}$H$_{22}$N$_2$O$_4$: *m/e* 413(M + Na); found: 413.

N,N'-(Furan-2-ylmethylene) dibenzamide (3n). Yellowish solid, m.p.: 206-208 °C; IR (KBr): ν$_{max}$ = 3277,

2928, 1650, 1489, 1340, 1273, 1137, 1051, 800, 706 cm^{-1}. ^{1}H NMR (500.1 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 7.03 (1H, t, *J* = 8.2 Hz, CH), 7.27-7.51 (9H, m, aroma), 7.87 (4H, d, *J* = 7.7 Hz, CH), 8.97 (2H, d, *J* = 6.9 Hz, NH). ^{13}C NMR (125.7 MHz, DMSO-d_6, Me$_4$Si): δ (ppm): 165.7, 152.7, 144.3, 140.7, 134.3, 130.1, 128.8, 127.9, 126.9, 119.2, 59.1. FAB Mass calcd. for C$_{19}$H$_{16}$N$_2$O$_3$: *m/e* 343 (M + Na); found: 343.

RESULTS AND DISCUSSION

Initially the reaction of benzaldehyde **1a** with benzamide **2a** (2 equivalents) and catalytic amount of boric acid in anhydrous toluene was refluxed. After completion of the reaction followed by standard work-up and purification *N,N'*-alkylidene bisamides **3a** was obtained in 92% yield (Scheme 1). To understand the effect of solvent, the reaction has been carried out with various solvents (Table 1, entries 1-6). No product was observed in CH$_3$CN and DCM as solvents (Table 1, entries 1 and 5). However, a considerable amount of desired products were formed in EtOH, EtOAc and THF (Table 1, entries 2, 3 and 6). Best yield was obtained when the reaction was performed in refluxing toluene with lesser reaction time and found as optimum condition (Table 1, entry 4).

The results indicated that reactions at elevated temperature (120 ℃) afforded maximum yield with shorter reaction time. It should be noted that in the absence of boric acid catalyst, only a trace of desired product was obtained due to equilibrium and the reaction takes longer time to complete (Table 1, entries 7). In order to reduce the reaction time, the
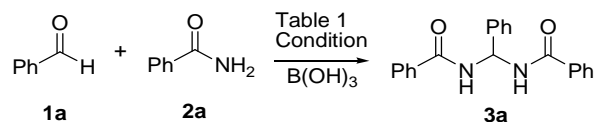
Scheme 1. Optimization of *N,N'*-alkylidene bisamides synthesis

reaction was carried out under microwave irradiation condition to afford the desired compound in 85% yield (Table 1, entry 8). The structure of the product **3a** was assigned based on the analysis of spectroscopic data such as FTIR, ^{1}H and ^{13}C NMR and mass spectroscopy.

With the optimal conditions in hand and to show the generality of the reaction, we examined the reaction of a series of aromatic aldehydes **1a-m** with benzamide **2a** and acetamide **2b** to afford symmetrical *N,N'*-alkylidene bisamides **3b-n** in moderate to good yields. The reaction is outlined in Scheme 2 and the results are summarized in Table 2.

Reactions are preceded well with various aromatic aldehydes and benzamide to provide symmetrical *N,N'*-alkylidene bisamides. However, the reaction of benzaldehyde with acetamide did not occur and only starting material was recovered in quantitative yield (Table 2, entry 2). In order to synthesize unsymmetrical *N,N'*-alkylidene bisamides, the reaction of benzaldehyde with one equivalent of benzamide and one equivalent of acetamide was carried out. The reaction provided only product of symmetrical *N,N'*-alkylidene

Table 1. Optimization Studies for the Synthesis of *N,N'*-Alkylidene Bisamides Using Boric Acid[a]

Entry	Solvent	Temp (°C)	Time(h)	Yield (%)[b]
1	CH$_3$CN	80	24	0
2	EtOH	80	20	35
3	EtOAc	80	25	45
4	Toluene	115	30	92
5	Dichloromethane	45	24	0
6	THF	65	28	30
7	Toluene (absence of Boric acid)	120	24	25
8	Microwave irradiation	80% PL	0.20	85

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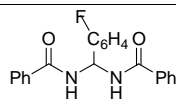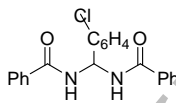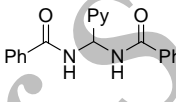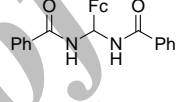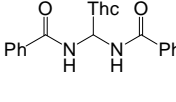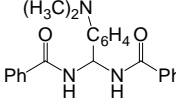Boric acid Catalyzed Efficient Synthesis of Symmetrical *N,N'*-Alkylidene Bisamides
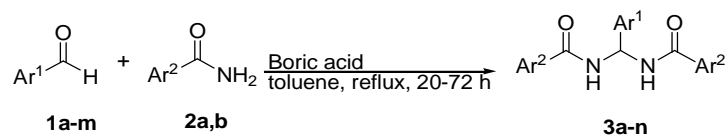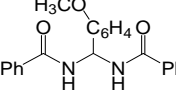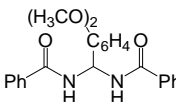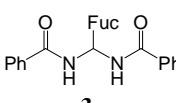

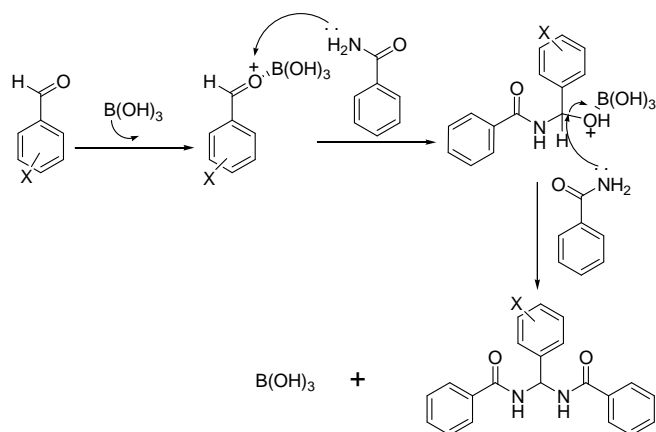*Scheme 2*. Synthesis of various *N,N'*-alkylidene bisamides

Table 2. Synthesis of *N,N'*-Alkylidene Bisamides Using Boric Acid under Classical Heating Conditions or Using Microwave Irradiation

Entry	Aldehyde	Amide	Product	Conventional heating		Microwave irradiation	
				Time (h)	Yield (%)	Time (min)	Yield (%)
1	C_6H_5CHO **1a**	$C_6H_5CONH_2$ **2a**	**3a**	16	92	20	85
2	C_6H_5CHO **1a**	CH_3CONH_2 **2b**	**3b**	24	0	40	0
3	C_6H_5CHO **1a**	$C_6H_5CONH_2 +$ **2a** CH_3CONH_2 **2b**	**3a**	24	38	40	35
4	4-Methyl benzaldehyde **1b**	$C_6H_5CONH_2$ **2a**	**3c**	60	80	25	75
5	4-Cyano benzaldehyde **1c**	$C_6H_5CONH_2$ **2a**	**3d**	30	68	35	64
6	4-Nitro benzaldehyde **1d**	$C_6H_5CONH_2$ **2a**	**3e**	30	65	30	60

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Table 2. Continued

7	4-Fluro benzaldehyde 1e	$C_6H_5CONH_2$ 2a	3f	30	55	35	50
8	4-Chloro benzaldehyde 1f	$C_6H_5CONH_2$ 2a	3g	48	65	30	60
9	Pyridine-4-aldehyde 1g	$C_6H_5CONH_2$ 2a	3h	45	70	35	64
10	Ferrocenealdehyde 1h	$C_6H_5CONH_2$ 2a	3i	40	65	40	58
11	Anisaldehyde 1i	$C_6H_5CONH_2$ 2a	3j	45	78	25	73
12	Thiophene-2-carboxyaldehyde 1j	$C_6H_5CONH_2$ 2a	3k	32	80	25	77
13	Dimethyl-Aminobenzaldehyde 1k	$C_6H_5CONH_2$ 2a	3l	70	38	35	32
14	3,4-Dimethoxy benzaldehyde 1l	$C_6H_5CONH_2$ 2a	3m	65	45	25	40
15	Furan-2-carboxyaldehyde 1m	$C_6H_5CONH_2$ 2a	3n	40	60	35	54

Scheme 3. A plausible reaction mechanism for the synthesis of symmetrical *N,N'*-alkylidene bisamides

bisamides in 38% yield with quantitative amount of unreacted acetamide (Table 2, entry 3). The reaction of 4-pyridylaldehyde under optimized condition afforded excellent yield of pyridine substituted *N,N'*-alkylidene bisamides (Table 2, entry 9). It appears that substitution at the 4-position of aromatic aldehyde produced no significant effects; aldehydes possessing electron-donating groups or amide bearing electron-withdrawing groups caused slightly lower yields. Interestingly, the reaction of ferrocenealdehyde with benzamide proceeds well and gave the desired product in good yield (Table 2, entry 10). The reaction in Dean-Stark trap apparatus has been carried out and found the reduction of reaction time without altering the yield with lesser reaction time (1.5 h). More to reduce the reaction time, all the reactions reported in (Table 2) were also carried out under neat microwave irradiation condition and found to afford almost same yields of the products as that of thermal reaction within 15-40 min.

A plausible reaction mechanism for the synthesis of symmetrical *N,N'*-alkylidene bisamides under boric acid as a Lewis acid is shown in Scheme 3.

In conclusion, boric acid has been successfully used as effective catalyst for the synthesis of symmetrical *N,N'*-alkylidene bisamides for the first time. This procedure has advantages in competition with the previously reported methods, in terms of yield, green catalyst, mild reaction condition, simple procedure, lack of toxicity, low cost, the use of a commercially available catalyst and simplicity of work-up. Further work using the catalyst for the synthesis of novel heterocycles is in progress in this laboratory.

ACKNOWLEDGEMENTS

REFERENCES

[1] T. Yamazaki, K.I. Nunami, M. Goodman, Biopolymers 31 (1991) 1513.

[2] M. Goodman, H. Shao, Pure & Appl. Chem. 68 (1996) 1303.

[3] W.A. Noyes, D.B. Forman, J. Am. Chem. Soc. 55 (1933) 3493.

[4] N.O. Brace, G.J. Mantell, J. Org. Chem. 26 (1961) 5176.

[5] E.E. Magat, B.F. Faris, J.E. Reith, L.F. Salisbury, J. Am. Chem. Soc. 73 (1951) 1028.

[6] A.H. Fernandez, R.M. Alvarez, T.M. Abajo, Synthesis (1996) 1299.

[7] S. Zhu, G. Xu, Q. Chu, Y. Xu, C. Qui, J. Fluorine Chem. 93 (1999) 69.

[8] N.P. Selvam, S. Saranya, P.T. Perumal, Can. J. Chem. 85 (2008) 32.

[9] A. Milenkovic, F. Fache, R. Faure, M. Lemaire, Synth. Commun. 29 (1999) 1535.

[10] S.I. Zav'yalov, L.B. Kulikova, Khimiko-Farmatsevticheskii Zhurnal 26 (1992) 88. CAN 117:150650.

[11] G.S. Bhatnagar, K.C. Pandya, Proceedings-Indian Academy of Sciences, Section A 24 (1946) 487. CAN 41:18893; b) G.S. Bhatnagar, K.C. Pandya, Proceedings-Indian Academy of Sciences, Section A 10A (1939) 285.

[12] M.K. Chaudhuri, S. Hussain, M. Lakshmi Kantam, B.

Neelima, Tetrahedron Lett. 46 (2005) 8329.

[13] M.K. Chaudhuri, S. Hussain, J. Mol. Cat. A 269 (2007) 214.

[14] S.J. Tu, X.T. Zhu, F. Fang, X.J. Zhang, S.L. Zhu, T.J. Li, D.Q. Shi, X.S. Wang, S.J. Ji, Tetrahedron Lett. 44 (2003) 6153.

[15] G.C.M. Kondaiah, L. Amarnath Reddy, K. Srihari Babu, V.M. Gurav, K.G. Huge, R. Bandichhor, P. Pratap Reddy, A. Bhattacharya, R. Vijaya Ananda, Tetrahedron Lett. 49 (2008) 106.

[16] J. Pernak, B. Mrowczynski, J. Weglewski, Synthesis 12 (1994) 1415.

[17] S.S. Rawalay, C.F. Beam, J. Chem. Eng. Data 27

(1982) 475.

[18] L. Hazai, G. Deak, Acta Chim. Hung. 121 (1986) 237.

[19] a) A. Kumar, R.A. Maurya, Tetrahedron Lett. 49 (2008) 5471; b) T. Pingwah, Org. Synth. 81(2005) 262; c) K.F. Shelke, S.B. Sapkal, S.S. Sonar, B.R. Madje, B.B. Shingate, M.S. Shingare, Bull. Korean Chem. Soc. 30 (2009) 1057.

[20] P.V. Shinde, S.S. Sonar, B.B. Shingate, M.S. Shingare, Tetrahedron Lett. 51 (2010) 1308.

[21] D.D. Perrin, W.F.L. Armearego, Purification of Laboratory Chemicals, 3rd ed., Pergamon Press, New York, 1988.

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