One-Pot Three-Component Synthesis of Dihydrobenzo- and Naphtho[*e*]-1,3-oxazines in Water

Bijoy P. Mathew and Mahendra Nath*

Department of Chemistry, University of Delhi, Delhi-110 007, India
*E-mail: mnath@chemistry.du.ac.in

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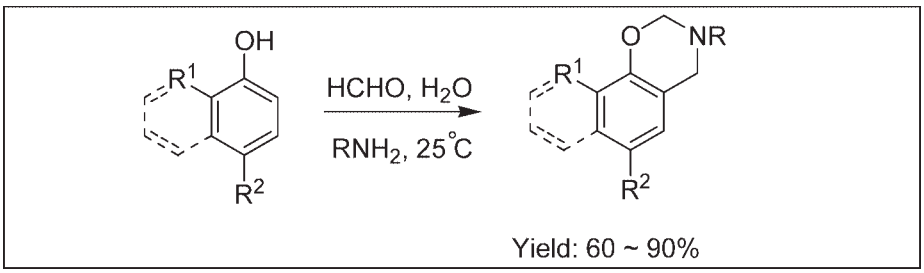

A simple, green and efficient method has been developed for the synthesis of biologically and materially important dihydrobenzo/naphtho[*e*]-1,3-oxazines in good to excellent yields through a Mannich-type condensation cyclization reaction of aromatic alcohols with HCHO and primary amines in aqueous media at ambient temperature.

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INTRODUCTION

Recently, great attention has been focused to develop nonhazardous and environment friendly synthetic strategies for organic reactions by replacing volatile organic solvents with nontoxic and noninflammable media such as water, ionic liquids, supercritical fluids, and even under neat conditions [1]. Among these, water has emerged as a versatile solvent for several organic transformations [2]. Water is not only a desirable solvent for chemical processes because of cost, safety, and environmental concerns but also provides completely new reactivity. The wide range of organic reactions is known in water including Diels-Alder cycloadditions, Knoevenagal condensations, Aldol-condensations, epoxidation, and oxidation–reduction reactions [3–7]. In addition to these, water has also been used as a solvent in reactions involving carbanions, carbocations, radicals, carbenes, and most recently transition metal catalyzed reactions [8]. In many organic reactions, the use of water as a solvent not only accelerates the rate and the yield but also enhances enantioselectivity in chiral synthesis [9].

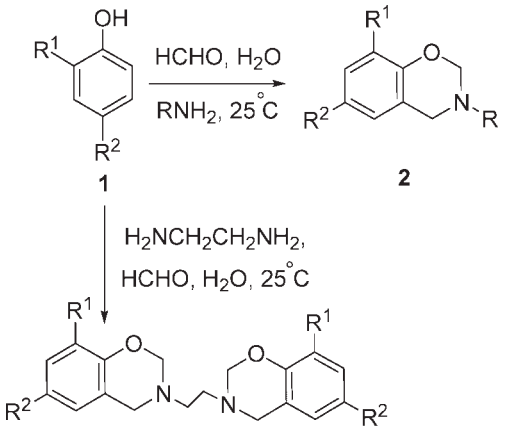
Dihydrobenzo/naphtho[*e*]-1,3-oxazines are known to exhibit a wide range of valuable pharmacological properties as antitumor [10], antibacterial [11], anti-HIV [12], and antimicrobial agents [13]. In addition, 6-arylbenzoxazines have also been examined as progesterone receptor modulators and reported as potent nonsteroidal progesterone receptor agonists [14]. On the other hand, these molecules are materially very useful for making phenol type polymers such as polybenzoxazines, offers lucrative mechanical, and electrical properties [15]. The diverse applications of this class of heterocyclic systems inspired chemists to develop new and efficient synthetic strategies for the preparation of these bioactive and materially important molecules.

The syntheses of dihydro-1,3-oxazines have been previously reported by several investigators *via* Mannich-type condensation of a phenol or naphthol with formaldehyde and primary amine using highly inflammable organic solvents [16] and alkaline media [17]. Further, researchers have also been successful in developing mild and solvent free protocol for the construction of these molecules [18]. However, many of these processes are associated with several shortcomings such as long duration of reaction, high temperature, use of volatile and hazardous organic solvents, and occurrence of side products. As a part of our research program to develop a simple methodology for the preparation of target compounds of pharmaceutical interests, we wish to report herein a simple one-pot three-component synthesis of 3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazines (Scheme 1) and dihydronaphtho[*e*]-1,3-oxazines (Scheme 2) in neat water at ambient temperature.

RESULTS AND DISCUSSION

Our initial efforts are focused on the search of a green solvent to carry out the synthesis of dihydrobenzo/naphtho[*e*]-1,3-oxazines at ambient temperature. In this connection, water was chosen for the condensation reaction of phenol, HCHO, and butylamine at room temperature. The completion of the reaction was monitored by TLC

Scheme 1



and the product was extracted with ethyl acetate, and purified by column chromatography on silica gel using ethyl acetate/heptane as eluent to afford 3-butyl-6-chloro-3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazine (**2a**) in 64% yield. To demonstrate the versatility of this methodology, synthesis of a variety of 3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazines (**2b-g**) and 1,2-bis(3,4-dihydro-2*H*-benzo [*e*]-1,3-oxazin-3-yl)ethanes (**3a-d**) has been carried out in good to excellent yields (Table 1).

Under these optimized reaction conditions, the scope of the reaction was then further explored by reacting 1-naphthol (**4a**) and 2-naphthol (**4b**) with aqueous HCHO and primary amines in water at ambient temperature separately to obtain 3,4-dihydro-2*H*-naphtho[*e*]-1,3-oxazines (**5a-d**) and 2,3-dihydro-1*H*-naphtho[*e*]-1,3-oxazines (**6a-g**), respectively in good yields (Table 2).

Our synthetic strategy does not require hazardous and toxic organic solvents, and the products were isolated by extraction with ethyl acetate, a preferred and safer solvent [19]. In fact, the dihydrobenzo/naphtho[*e*]-1,3-oxazines were prepared in a short time (30 min to 1 h) at 25°C in water, whereas in other organic solvents the reaction takes several hours to days for complete consumption of starting materials. All the synthesized dihydro-1,3-oxazine derivatives have been characterized by ^{1}H, ^{13}C NMR, IR, mass, and elemental analyses. The physical data of all the known compounds (**2a-c, 3a, b, d, 5a-c, 6a-e**) are in agreement with those of reported data [11,16,20,21]. The spectral and analytical data of new compounds (**2d-g, 3c, 5d, 6f-g**) are presented in the experimental section.

Furthermore, the polymerization behavior of novel benzoxazine analogues (**2f**; 4.79 mg & **3c**; 6.30 mg) was examined by Differential Scanning Calorimetry (DSC) with a heating rate of 10°C per min. The endothermic peaks at 67.5°C and 167°C are attributed to the melting of both **2f** and **3c**. For 6-chloro-3-(2-chlorophenyl)-3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazine (**2f**), an exotherm was observed due to the ring opening polymerization [16j] with onset at 267°C and maximum at 270°C. The amount of heat of polymerization is 218 J/g. The DSC trace of 1,2-bis(8-chloro-2*H*-benzo[*e*]-1,3-oxazin-3(4*H*)-yl)ethane (**3c**) showed an exotherm with onset

Table 1

Synthesis of 3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazines and 1,2-bis(2*H*-benzo[*e*]-1,3-oxazin-3(4*H*)-yl)ethanes in water.

Entry	Compound	R^1	R^2	R	Yield (%)
1	**2a**	H	H	C_4H_9	64
2	**2b**	H	H	C_6H_5	77
3	**2c**	H	H	C_6H_{11}	80
4	**2d**	Cl	H	4–$CH_3C_6H_4$	90
5	**2e**	H	CH_3	2–ClC_6H_4	82
6	**2f**	H	Cl	2–ClC_6H_4	84
7	**2g**	Cl	H	2–ClC_6H_4	74
8	**3a**	H	H	–	87
9	**3b**	H	Cl	–	75
10	**3c**	Cl	H	–	77
11	**3d**	Cl	Cl	–	70

Scheme 2

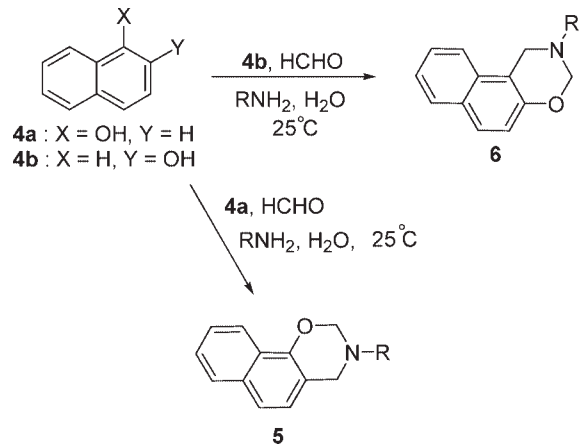


Table 2

Synthesis of 3,4-dihydro-2*H*-naphtho[*e*]-1,3-oxazines and 2,3-dihydro-1H-naphtho[*e*]-1,3-oxazines in water.

Entry	Compound	R	Yield (%)
1	**5a**	C_6H_5	90
2	**5b**	$CH_2C_6H_5$	60
3	**5c**	C_6H_{11}	62
4	**5d**	C_4H_9	85
5	**6a**	C_6H_5	79
6	**6b**	$CH_2C_6H_5$	75
7	**6c**	C_6H_{11}	87
8	**6d**	C_4H_9	80
9	**6e**	4-BrC_6H_4	73
10	**6f**	2,5-$Cl_2C_6H_3$	65
11	**6g**	2,4,6-$Cl_3C_6H_2$	63

at 195°C and maximum at 200°C. The amount of heat of polymerization for **3c** is 289 J/g. The shift of the cure exotherm to a lower temperature in case of **3c** than that of **2f** is possibly due to the flexibilizing effect of the ethylene bridge, which accelerated the ring opening polymerization.

In conclusion, we have developed a rapid, energy efficient, and environmentally conducive one-pot process for the preparation of dihydrobenzo- and naphtho[*e*]-1,3-oxazines in good yields at ambient temperature. In comparison with reported methods for making these molecules, the procedure presented herein avoids the use of hazardous organic media and the reactions are run in pure water, which is the cheapest and most harmless solvent available. Finally, these green advantages make the process more attractive for the synthesis of various biologically important dihydro-1,3-oxazine molecules.

EXPERIMENTAL

All the chemicals were used as received without any further purification. The products were purified by column chromatography using silica gel (60–120 mesh size). ^{1}H and ^{13}C NMR were recorded on Brucker 300 MHz spectrometer in $CDCl_3$ and referenced to the proton or carbon resonances resulting from incomplete deuteration of deuterated solvent. IR spectra were recorded on Perkin Elmer IR spectrometer and mass spectra were recorded on ESI-MS (micromass LCT, Water) mass spectrometer. DSC traces were recorded by using 4–6 mg samples in aluminium pan on Perkin Elmer DSC with a heating rate of 10°C min^{-1} in nitrogen atmosphere. Elemental Analyses were obtained from CHNSO Lab, University Science Instrumentation Centre, University of Delhi, Delhi.

General procedure for the one-pot synthesis of dihydrobenzo/naphtho[*e*]-1,3-oxazines (2a-g, 5a-d, 6a-g). To a mixture of aromatic alcohol (10 mmol) and primary amine (10 mmol) in water (20 ml), formalin (37%, w/v, 20 mmol) was added. The reaction mixture was stirred at 25°C for 30 min to 1 h. After completion of the reaction, the product was extracted with ethyl acetate (20 ml, two times). The organic layers were combined and washed with 10% aqueous NaOH solution (30 ml, two times) followed by water (50 ml). The organic layer was dried over sodium sulfate and evaporated under reduced pressure. The crude product was purified by column chromatography on silica gel using ethyl acetate/heptane as eluent to afford the desired product.

8-Chloro-3-(4-methylphenyl)-3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazine (2d). Brown colored viscous liquid; ν_{max} (film, cm^{-1}): 2922, 2851, 1615, 1574, 1515, 1461, 1378, 1240, 927, 818; ^{1}H nmr ($CDCl_3$): δ 2.26 (s, 3H, CH_3), 4.59 (s, 2H, NCH_2), 5.44 (s, 2H, OCH_2N), 6.79 (dd, J = 7.8, 7.6 Hz, 1H, ArH), 6.91 (d, J = 7.6 Hz, 1H, ArH), 7.02 (d, J = 8.3 Hz, 2H, ArH), 7.07 (d, J = 8.3 Hz, 2H, ArH), 7.19 (d, J = 7.5 Hz, 1H, ArH), HRMS: exact mass calculated for $C_{15}H_{13}ClNO$ (M^+-H): 258.0686; found: 258.0246.

3-(2-Chlorophenyl)-6-methyl-3,4-dihydro-2*H*-benzo-[*e*]-1,3-oxazine (2e). Viscous liquid; ν_{max} (film, cm^{-1}): 2924, 2856, 1618, 1589, 1502, 1483, 1441, 1226, 1041, 971, 949, 916, 814; ^{1}H nmr ($CDCl_3$): δ 2.24 (s, 3H, CH_3), 4.52 (s, 2H, NCH_2), 5.25 (s, 2H, OCH_2N), 6.72–7.38 (m, 7H, ArH); ^{13}C nmr ($CDCl_3$): δ 152.2, 146.9, 130.9, 130.7, 128.9, 128.1, 127.4, 125.3, 123.3, 120.8, 117.1, 80.8, 51.3, 21.0. HRMS: exact mass calculated for $C_{15}H_{14}ClNO$ (M^+): 259.0764; found: 259.0696.

6-Chloro-3-(2-chlorophenyl)-3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazine (2f). Pale yellow solid; mp 67.5 °C; ν_{max} (film, cm^{-1}): 2921, 2851, 1588, 1481, 1439, 1413, 1248, 1228, 1160, 1040, 942, 878, 815; ^{1}H nmr ($CDCl_3$): δ 4.52 (s, 2H, NCH_2), 5.26 (s, 2H, OCH_2N), 6.77–7.39 (m, 7H, ArH); ^{13}C nmr ($CDCl_3$): δ 153.0, 146.4, 131.0, 128.9, 128.4, 128.1, 126.8, 126.1, 125.6, 123.2, 122.5, 118.7, 81.0, 51.0; HRMS: exact mass calculated for $C_{14}H_{12}Cl_2NO$ (MH^+): 280.0296; found: 280.0770; *Anal*. Calcd. for $C_{14}H_{11}Cl_2NO$: C, 60.02; H, 3.96; N, 5.00. Found: C, 60.07; H, 4.03; N, 5.04.

8-Chloro-3-(2-chlorophenyl)-3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazine (2g). Viscous liquid; ^{1}H nmr ($CDCl_3$): δ 4.58 (s, 2H, NCH_2), 5.41 (s, 2H, OCH_2N), 6.81–6.92 (m, 2H, ArH), 7.03 (t, J = 7.6 Hz, 1H, ArH), 7.15 (t, J = 8 Hz, 1H, ArH), 7.24 (d, J = 8.1 Hz, 1H, ArH), 7.33–7.40 (m, 2H, ArH); HRMS: exact mass calculated for $C_{14}H_{12}Cl_2NO$ (MH^+): 280.0296; found: 280.0972.

3-Butyl-3,4-dihydro-2*H*-naphtho[2,1-*e*]-1,3-oxazine (5d). Dark viscous liquid; ν_{max} (film, cm^{-1}): 2924, 2853, 1578, 1465, 1404, 1070, 913, 801; ^{1}H nmr ($CDCl_3$): δ 0.90 (t, J = 7.3 Hz, 3H, CH_3), 1.32–1.39 (m, 2H, CH_2), 1.56–1.61 (m, 2H, CH_2), 2.79 (t, J = 7.4 Hz, 2H, CH_2), 4.09 (s, 2H, NCH_2), 5.04 (s, 2H, OCH_2N), 7.05 (d, J = 8.3 Hz, 1H, ArH), 7.35 (d, J = 8.4 Hz, 1H, ArH), 7.41–7.47 (m, 2H, ArH), 7.73–7.76 (m, 1H, ArH), 8.11–8.14 (m, 1H, ArH); HRMS: exact mass calculated for $C_{16}H_{19}NO$ (M^+): 241.1467; found: 241.5292.

2-(2,5-Dichloro-phenyl)-2,3-dihydro-1*H*-naphtho[1,2-*e*]-1,3-oxazine (6f). Pale yellow solid; mp 139°C; ν_{max} (film, cm^{-1}): 2890, 1625, 1600, 1582, 1559, 1516, 1474, 1435, 1403, 1383, 1267, 1230, 1191, 1159, 1131, 1102, 1059, 1047, 1008, 981, 950, 917, 857, 810; ^{1}H nmr ($CDCl_3$): 4.88 (s, 2H, NCH_2), 5.32 (s, 2H, OCH_2N), 6.97 (dd, J = 2.4 Hz, 1H, ArH), 7.10 (d, J = 8.9 Hz, 1H, ArH), 7.29 (d, J = 8.5 Hz, 1H, ArH), 7.35–7.40 (m, 2H, ArH), 7.47–7.52 (m, 1H, ArH), 7.61–7.69 (m, 2H, ArH), 7.78 (d, J = 8.0 Hz, 1H, ArH); *Anal*. Calcd. for $C_{18}H_{13}Cl_2NO$: C, 65.47; H, 3.97, N, 4.24. Found: C, 65.19, H, 4.27, N, 4.16.

2-(2,4,6-Trichloro-phenyl)-2,3-dihydro-1*H*-naphtho[1,2-*e*]-1,3-oxazine (6g). Pale yellow solid; mp 122°C; ν_{max} (film, cm^{-1}): 2891, 1626, 1599, 1516, 1469, 1382, 1334, 1229, 1157, 1138, 1068, 1009, 953, 921, 880, 811; ^{1}H nmr ($CDCl_3$): δ 4.85 (s, 2H, NCH_2), 5.24 (s, 2H, OCH_2N), 7.09 (d, J = 8.9 Hz, 1H, ArH), 7.38 (t, J = 7.1 Hz, 1H, ArH), 7.46–7.52 (m, 3H, ArH), 7.61 (d, J = 8.3 Hz, 1H, ArH), 7.68 (d, J = 8.9 Hz, 1H, ArH), 7.78 (d, J = 8.0 Hz, 1H, ArH); *Anal*. Calcd. for $C_{18}H_{12}Cl_3NO$: C, 59.29, H, 3.32, N, 3.84. Found: C, 59.22, H, 3.57, N, 3.79.

General procedure for the one-pot synthesis of 1,2-bis(2*H*-benzo[*e*]-1,3-oxazin-3(4*H*)-yl)ethanes (3a-d). To a mixture of aromatic alcohol (20 mmol) and ethylene diamine (10 mmol) in water (20 ml), formalin (37%, w/v, 40 mmol) was added. The reaction mixture was stirred at 25°C for 30 min. After completion of the reaction, the product was extracted with ethyl acetate (20 ml, two times). The organic layers were combined and washed with 10% aqueous NaOH solution (30 ml, two times) followed by water (50 ml). The organic layer was dried over sodium sulfate and evaporated

under reduced pressure. The crude solid was washed with ethanol to obtained sufficiently pure product.

1,2-Bis(8-chloro-3,4-dihydro-2*H*-benzo[*e*]-1,3-oxazin-3(4*H*)-yl)ethane (3c). White solid; mp 167°C; ν_{max} (film, cm^{-1}) 2918, 2854, 1572, 1456, 1353, 1321, 1231, 1120, 1071, 1031, 981, 894, 762; ^1H nmr ($CDCl_3$): δ 2.98 (s, 4H, NCH_2CH_2N), 4.04 (s, 4H, $2NCH_2$), 5.02 (s, 4H, $2OCH_2N$), 6.77–6.87 (m, 4H, ArH), 7.21 (d, $J = 6.8$ Hz, 2H, ArH). *Anal*. Calcd. for $C_{18}H_{18}Cl_2N_2O_2$: C, 59.19, H, 4.97, N, 7.67. Found: C, 59.39, H, 5.22, N, 7.91.

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REFERENCES AND NOTES

[1] (a) Lindstron, U. M. Chem Rev 2002, 102, 2751; (b) Wasserscheid, P.; Welton, T., Eds. Ionic Liquids in Synthesis; Wiley-VCH: Weinheim, 2003; (c) Jessop, P. G.; Leitner, W., Eds. Chemical Synthesis Using Supercritical Fluids; Wiley-VCH: Weinheim, 1999; (d) Tanaka, K. Solvent-Free Organic Synthesis; Wiley-VCH: Weinheim, 2003; (e) Sheldon, R. A. Green Chem 2005, 7, 267.

[2] (a) Kobayashi, S.; Manabe, K. Acc Chem Res 2002, 35, 209; (b) Li, C.-J. Chem Rev 2005, 105, 3095.

[3] Narayan, S.; Muldoon, J.; Finn, M. G.; Fokin, V. V.; Kolb, H. C.; Sharpless, K. B. Angew Chem Int Ed Engl 2005, 44, 3275.

[4] Amantini, D.; Fringuelli, F.; Piermatti, O.; Pizzo, F.; Vaccaro, L. Green Chem 2001, 3, 229.

[5] Fringuelli, F.; Piermatti, O.; Pizzo, F. In Organic Synthesis in Water; Grieco, P. A., Ed.; Blackie Academie and Professional: London, 1998; p 223.

[6] Lindstrom, U. M.; Ding, R.; Hidestal, O. Chem Commun 2005, 1773.

[7] (a) Matsushita, M.; Kamata, K.; Yamaguchi, K.; Mizuno, N. J Am Chem Soc 2005, 127, 6632; (b) Schlatter, A.; Kundu, M. K.; Woggon, W.-D. Angew Chem Int Ed Engl 2004, 43, 6731.

[8] Li, C.-J.; Chen, L. Chem Soc Rev 2006, 35, 68.

[9] Ribe, S.; Wipf, P. Chem Commun 2001, 299.

[10] (a) Kuehne, M. E.; Konopke, E. A. J Med Chem 1962, 5, 257; (b) Kuehne, M. E.; Konopke, E. A.; Lamberd, B. F. J Med Chem 1962, 5, 281; (c) Chylinska, J. B.; Urbanski, T. J Med Chem 1963, 6, 484; (d) Hsu, L.-Y.; Lin, C.-H. Heterocycles 1996, 43, 2687.

[11] Chylinska, J. B.; Janowiec, M.; Urbanski, T. Br J Pharmacol 1971, 43, 649.

[12] (a) Pedersen, O. S.; Pedersen, E. B. Synthesis 2000, 479; (b) Cocuzza, A. J.; Chidester, D. R.; Cordova, B. C.; Jeffrey, S.; Parsons, R. L.; Bacheler, L. T.; Erickson-Viitanen, S.; Trainor, G. L.; Ko, S. S. Bioorg Med Chem Lett 2001, 11, 1177.

[13] (a) Latif, N.; Mishriky, N.; Massad, F. Aust J Chem 1982, 35, 1037; (b) Skoda, J.; Votruba, I.; Farkas, J. Biochem Pharmacol 1979, 28, 1837.

[14] Zhang, P.; Terefenko, E. A.; Fensome, A.; Zhang, Z.; Zhu, Y.; Cohen, J.; Winneker, R.; Wrobel, J.; Yardley, J. Bioorg Med Chem Lett 2002, 12, 787.

[15] (a) Agag, T.; Takeichi, T. Macromolecules 2003, 36, 6010; (b) Liu, Y.-L.; Yu, J.-M. J Polym Sci. Part A: Polym Chem 2006, 44, 1890.

[16] (a) Burke, W. J. J Am Chem Soc 1949, 71, 609; (b) Burke, W. J.; Kolbezen, M. J.; Stephens, C. W. J Am Chem Soc 1952, 74, 3601; (c) Burke, W. J.; Murdock, K. C.; Ec, G. J Am Chem Soc 1954, 76, 1677; (d) Burke, W. J.; Reynolds, R. J. J Am Chem Soc 1954, 76, 1291; (e) Burke, W. J.; Hammer, C. R.; Weatherbee, C. J Org Chem 1961, 26, 4403; (f) Fields, D. L.; Miller, J. B.; Reynolds, D. D. J Org Chem 1962, 27, 2749; (g) Rivera, A.; Gallo, G. I.; Gayon, M. E. Synth Commun 1994, 24, 2081; (h) Schmidt, C.; Thondorf, I.; Kolehmainen, E.; Bohmer, V.; Vogt, W.; Rissanen, K. Tetrahedron Lett 1998, 39, 8833; (i) Higham, C. S.; Dowling, D. P.; Shaw, J. L.; Cetin, A.; Ziegler, C. J.; Farrell, J. R. Tetrahedron Lett. 2006, 47, 4419; (j) Agag, T.; Takeichi, T. J Polym Sci Part A: Polym Chem 2007, 45, 1878; (k) Burke, W. J.; Smith R. P.; Weatherbee, C. J Am Chem Soc 1952, 74, 602.

[17] (a) Miura, S.; Kano, N. Jpn. Pat. 2,000,169,456 (2000); (b) Miura, S.; Kano, N. Chem Abstr 2000, 133, 30736b; (c) Burke, W. J.; Glennie, E. L. M.; Weatherbee, C. J Org Chem 1964, 29, 909; (d) Na, L.; Ling, L.; Fengyun, D.; Yang, Z. Zhanjie 2005, 26, 20.

[18] (a) Inoue, S.; Kim, R.; Hoshino, Y.; Honda, K. Chem Commun 2006, 1974. (b) Brunovska, Z.; Liu, J. P.; Ishida, H. Macromol Chem Phys 1999, 200, 1745; (c) Ishida, H. U.S. Pat. 5,543,516 (1996); (d) Ishida, H. Chem Abstr 1996, 124, 203404m; (e) Hossein, F.; Ali, S. Chemistry (Rajkot, India) 2003, 1, 151.

[19] Alfonsi, K.; Colberg, J.; Dunn, P. J.; Fevig, T.; Jennings, S.; Johnson, T. A.; Kleine, H. P.; Knight, C.; Nagy, M. A.; Perry, D. A.; Stefaniak, M. Green Chem 2008, 10, 31.

[20] (a) Katritzky, A. R.; Xu, Y.-J.; Jain, R. J Org Chem 2002, 67, 8234; (b) Kiskan, B., Yagci, Y.; Sahmetlioglu, E.; Toppare, L. J Polym Sci Part A: Polym Chem 2007, 45, 999; (c) Wang, Y.-X.; Ishida, H. J Appl Polym Sci 2002, 86, 2953.

[21] (a) Agag, T. J Appl Polym Sci 2006, 100, 3769; (b) Rincon, J. M. Rev Colomb Quim 1977, 7, 57.