Indian Journal of Chemistry
Vol. 46B, October 2007, pp. 1672-1678

SDS/CH_2(CN)EWG/H_2O: An efficient and green system for the cleavage of nitrones and imines in water

Saikat Das Sharma, Pranjal Gogoi & Dilip Konwar*

Synthetic Organic Chemistry Division, North-East Institute of Science & Technology
(Formerly Regional Research Laboratory), Jorhat 785 006, India

E-mail: dkonwar@yahoo.co.uk

Received 18 September 2006; accepted (revised) 3 July 2007

SDS/CH_2(CN)EWG/H_2O system (EWG= electron withdrawing group) reacted with nitrones and imines in water to produce addition-elimination products with excellent yields at room temperature.

Keywords: SDS, imines, nitrones, green, cleavage, water

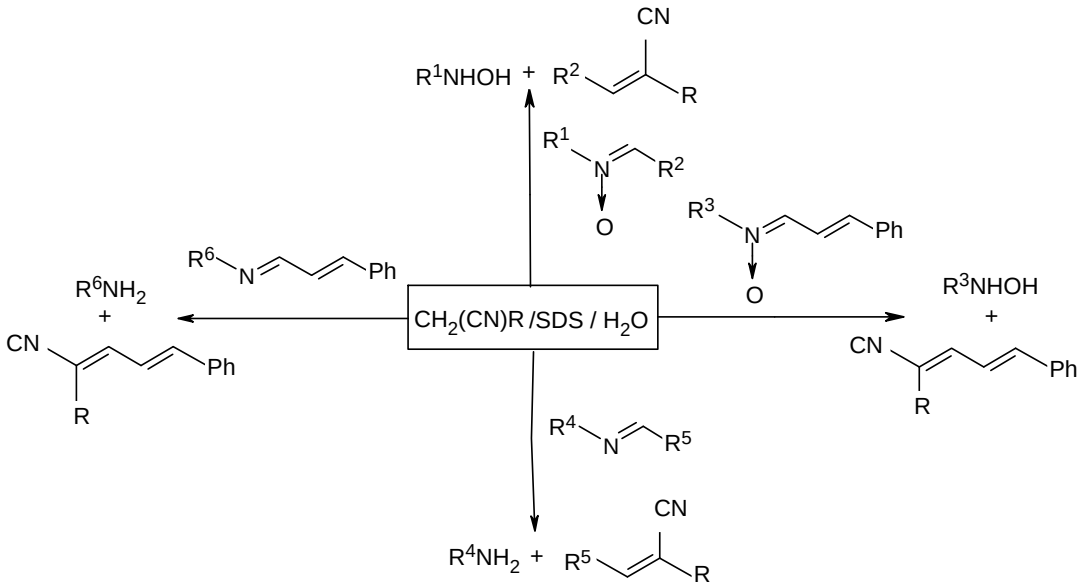
The development of environmentally benign chemical processes or methodologies have received much attention in recent years, and water as a solvent fulfils a major requirement because it is readily available, cheap and environmentally benign[1]. On the other hand, organic N-oxides are important compounds with a broad synthetic potential. They are useful as starting materials for the synthesis of biologically important compounds like amino acids[2], amino alcohols[3], β-lactams[4], carbohydrates[5], alkaloids[6] and isoxazoles[7]. In addition, nitrones undergo inter and intra-molecular 1,3-dipolar cycloadditions to various types of dipolarophiles like C=C, C=N, *etc*. producing mostly five membered heterocycles[8]. Besides, due to the appreciable polarity of the dipolarophile C=N bonds they are easily cleaved with hydrazines, hydroxyl amines, carbanions, *etc*.[9] Similarly, the C=N bonds of imines are cleaved with various acids[10], oxidizing agents like ozone[11], $KMnO_4$/alumina[12], chromium reagents[13], polymer supported CrO_3 (Ref. 14), $(PhSeO)_2O$ (Ref. 15), and cobalt carbonyl[16]. However, there are various limitations to their general application: use of toxic and hazardous transition metals *i.e.* Mn (Ref. 12), Cr (Ref. 13b-e), Ti (Ref. 10a), use of strong Lewis[10b,c] and Brønsted acids[10e], low temperature[11,13a], longer reaction time[13b], low yield[15], use of bases and organic solvents, *etc*.

In recent years, surfactants[17] have been used as synthetic reagent or catalyst[18] due to their inherent properties of low toxicity, easy handling and amphiphilic nature in a reaction medium. In this endeavor, a number of reactions like ester formation[18], aldol condensation[19], synthesis of nitrone and their 1,3-dipolar cycloaddition reactions[20] are performed successfully in the presence of surfactants in aqueous medium. In continuation of the research to develop synthetic methodologies in hydrated media[21], herein is reported for the first time, a green and efficient method for the cleavage of C=N bonds of nitrones and imines to give addition-elimination products by using SDS/CH_2(CN)EWG/H_2O system in water with excellent yields at RT (**Scheme I**).

Results and Discussion

To a stirred solution of nitrone (1 mmol), malononitrile (1 mmol) and sodium dodecyl sulphate (SDS, 0.5 mmol) were mixed in water (10 mL) and the reaction mixture was stirred for stipulated time (**Table I**) at 25-30°C. After work-up, synthetically important arylidinemalononitriles[22] and hydroxyl-amines were obtained in excellent yields. Similarly, imines gave arylidinemalononitriles and amines under the same reaction conditions in excellent yields (**Table II**). The cleavage of nitrones and imines by the system is applicable to unconjugated (entry **1a-g, 4a-c, 4g-j**), and conjugated (entry **1h-j, 4d-f**) nitrones (**Table I**) and imines (**Table II**). In case of nitrones, formation of isoxazoline[23] or Michael addition products[24] did not occur in the reaction mixtures. In all the cases it formed addition-elimination products in excellent yields. The temperature range of the reaction was determined to be 25-30°C.

The proposed mechanism envisages that the anionic part of the surfactant generates carbanion from malonitrile which attacks the benzylic carbon of a nitrone or imine to give addition-elimination



Scheme I

products and reforms the surfactant in the reaction mixture as shown below (**Scheme II**).

In conclusion, this is the first reported green and efficient method for the cleavage of nitrones and imines with SDS/CH_2(CN)EWG/H_2O system in water. The system does not involve acids, bases, transition metals and organic solvent in the reaction mixture. The yields of the reaction are excellent and the reaction can be conducted at 25-30°C.

Experimental Section

^{1}H NMR spectra were recorded on Avance DPX 300 MHz FT-NMR spectrometer. Chemical shifts are expressed in δ units relative to tetramethylsilane (TMS) signal as internal reference in $CDCl_3$. IR spectra were recorded on FTIR-system-2000 Perkin Elmer spectrometer in $CHCl_3$ or on KBr pellets. Mass spectra were recorded on Esquire 3000 mass spectrometer. Commercially available sodium dodecyl sulphate (SDS) was used as received. Nitrones and imines were prepared by following standard literature procedures. Column chromatography was performed over silica gel (60-120 mesh) using ethyl acetate/ hexane mixtures as eluent.

General procedure for cleavage of carbon-nitrogen double bond of imines and nitrones

In a 50 mL round-bottom flask, imine or nitrone (1 mmol), active methylene compound (1 mmol) and surfactant (sodium dodecyl sulphate, 0.5 mmol) in H_2O (10 mL) were added and stirred at RT. The progress of the reaction was monitored by TLC. After completion of the reaction, the product was extracted with ethyl acetate (2×25 mL), the organic layer washed with brine (2×15 mL), dried over anhyd. Na_2SO_4 and concentrated. The products were separated and purified by column chromatography over silica gel (60-120 mesh) using ethyl acetate/hexane mixtures as eluent. The physical and spectroscopic characterization data of all the compounds are in agreement with those of authentic samples[25].

Spectral characterization data of the aryl hydroxylamines obtained after the cleavage of carbon-nitrogen double bond of nitrones

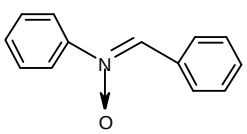
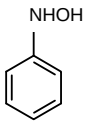
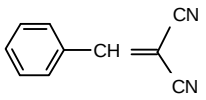
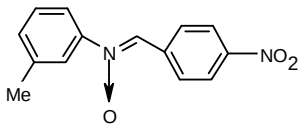
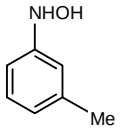
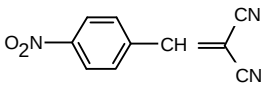
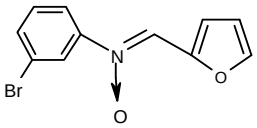
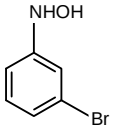
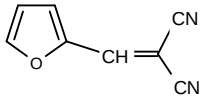
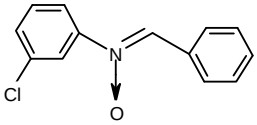
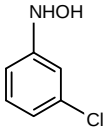
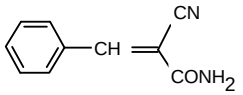
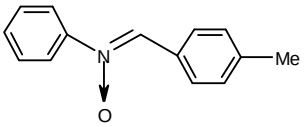
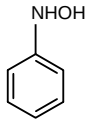
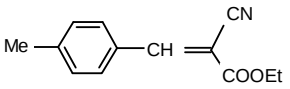
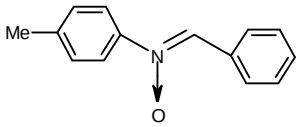
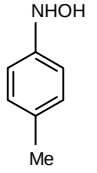
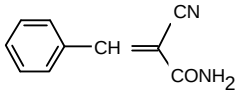
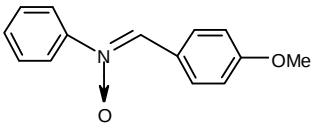
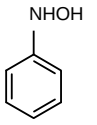
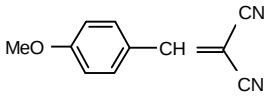
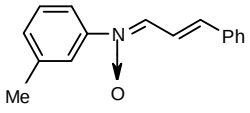
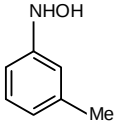
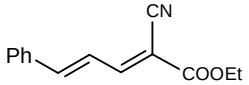
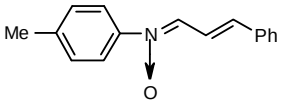
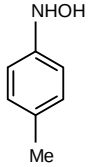
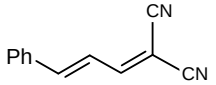
Phenylhydroxylamine, 2a: Yield 0.092 g (85%). m.p. 81-82ºC; ^{1}H NMR (DMSO-d_6): δ 8.2 (s, 1H), 8.0 (s, 1H), 6.7-7.0 (m, 5H); FT-IR (KBr): 3371.0, 3323.2 cm^{-1}; MS: *m/z* 109.14 (M^+).

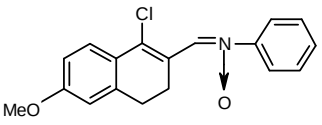
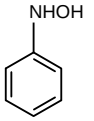
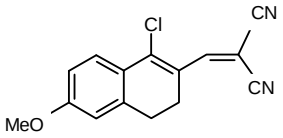
***m*-Toluylhydroxylamine, 2b**: Yield 0.102 g (83%). m.p. 67-68ºC; ^{1}H NMR (DMSO-d_6): δ 8.2 (s, 1H), 8.0 (s, 1H), 6.8-7.1 (m, 4H), 2.2 (s, 3H); FT-IR (KBr): 3370.7, 3325.2 cm^{-1}; MS: *m/z* 123.19 (M^+).

***m*-Bromophenylhydroxylamine, 2c**: Yield 0.152 g (81%). m.p. 66-67ºC; ^{1}H NMR (DMSO-d_6): δ 8.3 (s, 1H), 8.2 (s, 1H), 6.7-7.2 (m, 4H); FT-IR (KBr): 3372.3, 3328.5 cm^{-1}; MS: *m/z* 188.09 (M^+).

Indian Journal of Chemistry
Vol. 46B, October 2007, pp. 1672-1678

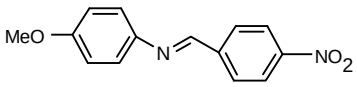
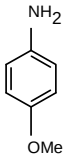
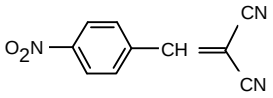
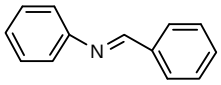
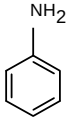
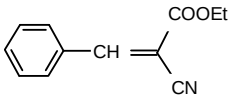
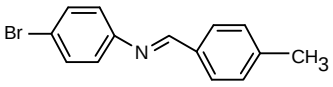
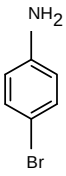
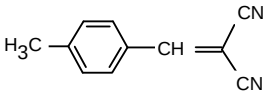
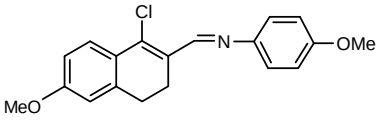
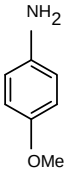
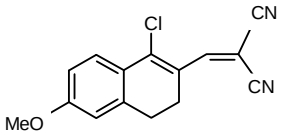
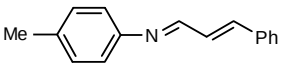
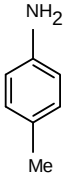
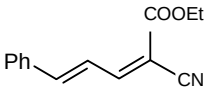
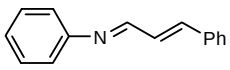
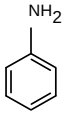
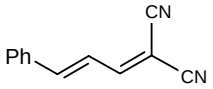
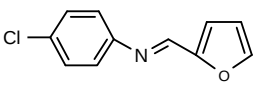
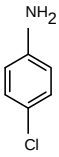
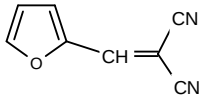
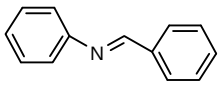
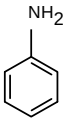
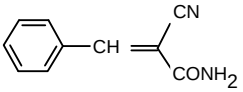
Table I — Reaction of nitrones **1a-j** with SDS/CH_2(CN)EWG/H_2O system

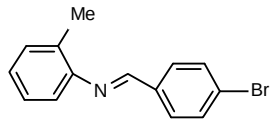
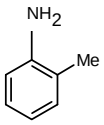
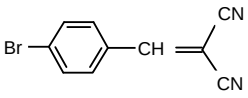
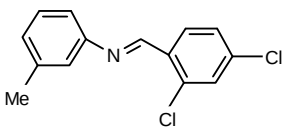
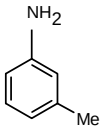
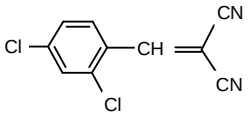
Entry	Substrate **1a-j**	Hydroxyl-amine **2a-j**	Yield (%)[a]	Arylidenemalononitrile **3a-j**	Yield (%)[a]	Time (min)
a			85		87	12
b			83		90	10
c			81		84	20
d			78		80	15
e			82		84	15
f			85		83	13
g			84		85	20
h			86		85	15
i			81		83	15

j			85		89	15

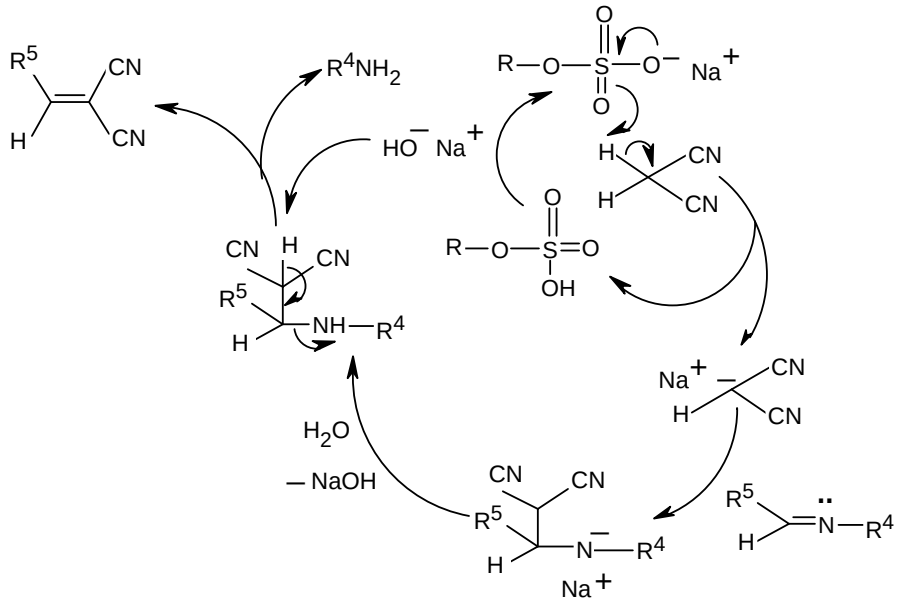
[a] Isolated yield.
[b] Stirring at 25-30°C.
[c] The structures of the compounds were determined by using ^{1}H NMR, FT-IR and mass spectroscopy and comparison with authentic samples[25].

Table II — Reaction of imines **4a-j** with SDS/CH_2(CN)EWG/H_2O system

Entry	Substrate **4a-j**	Amine **5a-j**	Yield (%)[a]	Arylidenemalononitrile **6a-j**	Yield (%)[a]	Time (min)
a			88		89	12
b			88		85	15
c			86		87	12
d			89		90	10
e			86		84	20
f			85		85	17
g			85		88	15
h			84		83	17

i			84		88	15
j			86		88	12

[a] Isolated yield.
[b] Stirring at 25-30°C.
[c] The structures of the compounds were determined by using ^{1}H NMR, FT-IR and mass spectroscopy and comparison with authentic samples[25].



Scheme II — Proposed mechanism for the cleavage of imines to arylidenemalononitriles and amines mediated by SDS/CH_2(CN)EWG/H_2O system

***m*-Chlorophenylhydroxylamine, 2d**: Yield 0.111g (78%). m.p. 50-51ºC; ^{1}H NMR (DMSO-d_6): δ 8.3 (s, 1H), 8.2 (s, 1H), 6.8-7.2 (m, 4H); FT-IR (KBr): 3371.4, 3328.9 cm^{-1}; MS: *m/z* 143.51 (M^+).

Phenylhydroxylamine, 2e: Yield 0.089 g (82%). m.p. 81-82ºC; ^{1}H NMR (DMSO-d_6): δ 8.2 (s, 1H), 8.0 (s, 1H), 6.7-7.1 (m, 5H); FT-IR (KBr): 3370.8, 3323.6 cm^{-1}; MS: *m/z* 109.14 (M^+).

***p*-Toluylhydroxylamine, 2f**: Yield 0.104 g (85%). m.p. 93-94ºC; ^{1}H NMR (DMSO-d_6): δ 8.2 (s, 1H), 8.0 (s, 1H), 6.8-7.0 (m, 4H), 2.2 (s, 3H); FT-IR (KBr): 3370.1, 3324.7 cm^{-1}; MS: *m/z* 123.09 (M^+).

Phenylhydroxylamine, 2g: Yield 0.091 g (84%). m.p. 81-82ºC; ^{1}H NMR (DMSO-d_6): δ 8.2 (s, 1H), 8.0 (s, 1H), 6.7-7.0 (m, 5H); FT-IR (KBr): 3370.5, 3323.9 cm^{-1}; MS: *m/z* 109.13 (M^+).

***m*-Toluylhydroxylamine, 2h**: Yield 0.105 g (86%). m.p. 67-68ºC; ^{1}H NMR (DMSO-d_6): δ 8.2 (s, 1H), 8.0 (s, 1H), 6.7-7.1 (m, 4H), 2.2 (s, 3H); FT-IR (KBr): 3370.4, 3324.9 cm^{-1}; MS: *m/z* 123.23 (M^+).

***p*-Toluylhydroxylamine, 2i**: Yield 0.099 g (81%). m.p. 93-94ºC; ^{1}H NMR (DMSO-d_6): δ 8.2 (s, 1H), 8.0 (s, 1H), 6.8-7.1 (m, 4H), 2.2 (s, 3H); FT-IR (KBr): 3371.2, 3325.7 cm^{-1}; MS: *m/z* 123.17 (M^+).

Phenylhydroxylamine, 2j: Yield 0.092 g (85%). m.p. 81-82ºC; ^{1}H NMR (DMSO-d_6): δ 8.2 (s, 1H), 8.0 (s, 1H), 6.8-7.1 (m, 5H); FT-IR (KBr): 3371.5, 3325.0 cm^{-1}; MS: *m/z* 109.09 (M^+).

Spectral characterization data of the arylidenemalononitriles obtained after the cleavage of carbon-nitrogen double bond of nitrones

Benzylidenemalonitrile, 3a: Yield 0.134 g (87%). m.p. 82-83ºC; ^{1}H NMR ($CDCl_3$): δ 7.27-8.15 (m, 6H); FT-IR (KBr): 2223.2, 1593.2 cm^{-1}; MS: *m/z* 154.11 (M^+).

(4-Nitrobenzylidene)malononitrile, 3b: Yield 0.179 g (90%). m.p. 159-60ºC; ^{1}H NMR ($CDCl_3$): δ 8.34-7.89 (m, 5H); FT-IR (KBr): 2232.4, 1580.7, 1522.8, 1345.9 cm^{-1}; MS: *m/z* 199.20 (M^+).

(2-Furanylmethylene)malononitrile, 3c: Yield 0.121 g (84%). m.p. 69-74ºC; ^{1}H NMR ($CDCl_3$): δ 7.3 (s), 7.55 (d, 1H), 7.15 (d, 1H), 6.50 (dd, 1H); FT-IR (KBr): 3124.8, 3042.0, 2231.4, 1606.4, 1529.5, 1021.7 cm^{-1}; MS: *m/z* 144.17 (M^+).

α-Cyanocinnamide, 3d: Yield 0.137 g (80%). m.p. 123-24ºC; ^{1}H NMR ($CDCl_3$) δ 8.25 (s, 1H), 7.12-8.12 (m, 7H); FT-IR (KBr): 3350.5, 2225.2,1680.0 cm^{-1}; MS: *m/z* 172.22 (M^+).

α-Cyano-4-methyl-ethylcinnamate, 3e: Yield 0.180 g (84%). m.p. 89-91ºC; ^{1}H NMR ($CDCl_3$): δ 7.97 (s, 1H), 7.7 (d, 2H), 7.18 (d, 2H), 4.19 (q, 2H), 2.30 (s, 3H), 1.30 (t, 3H); FT-IR (KBr): 2218.3, 1725.5, 1595.3 cm^{-1}; MS: *m/z* 215.19 (M^+).

α-Cyanocinnamide, 3f: Yield 0.142 g (83%). m.p. 123-24ºC; ^{1}H NMR ($CDCl_3$): δ 8.24 (s, 1H), 7.12-8.11 (m, 7H); FT-IR (KBr): 3350.1, 2225.5, 1680.1 cm^{-1}; MS: *m/z* 172.26 (M^+).

(4-Methoxybenzylidene)malononitrile, 3g: Yield 0.156 g (85%). m.p. 114-15ºC; ^{1}H NMR ($CDCl_3$): δ 8.11 (s, 1H), 8.05 (s, 1H), 7.2-7.5 (m), 3.7 (s, 3H); FT-IR (KBr): 2228.5, 1598.3 cm^{-1}; MS: *m/z* 184.15 (M^+).

2-Cyano-5-phenyl-ethylpentadienate, 3h: Yield 0.193 g (85%). m.p. 115-16ºC; ^{1}H NMR ($CDCl_3$): δ 7.2-7.6 (m, 8H), 4.2 (q, 2H), 1.3 (t, 3H); FT-IR (KBr): 2222.3, 1725.9, 1594.2 cm^{-1}; MS: *m/z* 227.29 (M^+).

Cinnamylidenemalononitrile, 3i: Yield 0.149 g (83%). m.p. 126-27ºC; ^{1}H NMR ($CDCl_3$): δ 7.0-7.7 (m, 8H); FT-IR (KBr): 2222.8, 1594.8 cm^{-1}; MS: *m/z* 180.26 (M^+).

1-Chloro-6-methoxy-2-[β, β-dicyano]-3,4-dihydro-naphthalene, 3j: Yield 0.241 g (89%). m.p. 190-91ºC; ^{1}H NMR ($CDCl_3$): δ 8.3 (s, 1H), 7.4 (d, 1H), 7.12 (d, 1H) 6.65 (s, 1H), 3.8 (s, 3H), 2.65-3.00 (m, 4H); FT-IR (KBr): 2219.7, 1611.9, 1517.9, 1282.6 cm^{-1}; MS: *m/z* 270.79 (M^+).

Spectral characterization data of the amines obtained after the cleavage of carbon-nitrogen double bond of imines

***p*-Anisidine, 5a**: Yield 0.108 g (88%). m.p. 59-60ºC; ^{1}H NMR ($CDCl_3$): δ 6.7 (d, 2H), 6.6 (d, 2H), 3.7 (s, 3H), 3.4 (s, 2H); FT-IR (KBr): 3351.9 cm^{-1}; MS: *m/z* 123.08 (M^+).

Aniline, 5b: Yield 0.081 g (88%). ^{1}H NMR ($CDCl_3$): δ 6.7-7.4 (m, 5H), 3.6 (s, 2H); FT-IR ($CHCl_3$): 3351.3 cm^{-1}; MS: *m/z* 93.01 (M^+).

***p*-Bromoaniline, 5c**: Yield 0.147 g (86%). m.p. 62-63ºC; ^{1}H NMR ($CDCl_3$): δ 6.5-7.4 (m, 4H), 3.6 (s, 2H); FT-IR ($CHCl_3$): 3351.8 cm^{-1}; MS: *m/z* 172.09 (M^+).

***p*-Anisidine, 5d**: Yield 0.109 g (89%). m.p. 59-60ºC; ^{1}H NMR ($CDCl_3$): δ 6.7 (d, 2H), 6.6 (d, 2H), 3.7 (s, 3H), 3.4 (s, 2H); FT-IR (KBr): 3351.7 cm^{-1}; MS: *m/z* 123.02 (M^+).

***p*-Toluidine, 5e**: Yield 0.092 g (86%). m.p. 43-44ºC; ^{1}H NMR ($CDCl_3$): δ 6.6 (d, 2H), 6.9 (d, 2H), 3.5 (s, 2H), 2.2 (s, 3H); FT-IR ($CHCl_3$): 3350.7 cm^{-1}; MS: *m/z* 107.10 (M^+).

Aniline, 5f: Yield 0.079 g (85%). ^{1}H NMR ($CDCl_3$): δ 6.7-7.3 (m, 5H), 3.6 (s, 2H); FT-IR ($CHCl_3$): 3351.7 cm^{-1}; MS: *m/z* 93.09 (M^+).

***p*-Chloroaniline, 5g**: Yield 0.108 g (85%). m.p. 72-73ºC; ^{1}H NMR ($CDCl_3$): δ 6.5-7.2 (m, 4H), 3.5 (s, 2H); FT-IR ($CHCl_3$): 3350.2 cm^{-1}; MS: *m/z* 127.49 (M^+).

Aniline, 5h: Yield 0.078 g (84%). ^{1}H NMR ($CDCl_3$): δ 6.8-7.3 (m, 5H), 3.6 (s, 2H); FT-IR ($CHCl_3$): 3350.8 cm^{-1}; MS: *m/z* 93.01 (M^+).

***o*-Toluidine, 5i**: Yield 0.090 g (84%). ^{1}H NMR ($CDCl_3$): δ 6.5-7.1 (m, 4H), 3.5 (s, 2H), 2.3 (s, 3H); FT-IR ($CHCl_3$): 3352.1 cm^{-1}; MS: *m/z* 107.19 (M^+).

***m*-Toluidine, 5j**: Yield 0.092 g (86%). ^{1}H NMR ($CDCl_3$): δ 6.5-7.0 (m, 4H), 3.6 (s, 2H), 2.3 (s, 3H); FT-IR ($CHCl_3$): 3350.3 cm^{-1}; MS: *m/z* 107.20 (M^+).

Spectral characterization data of the arylidenemalononitriles obtained after the cleavage of carbon-nitrogen double bond of imines

(4-Nitrobenzylidene)malononitrile, 6a: Yield 0.177 g (89%). m.p. 160-61ºC; ^{1}H NMR ($CDCl_3$): δ 8.3-7.7 (m, 5H); FT-IR (KBr): 2231.8, 1581.3, 1520.9, 1342.6 cm^{-1}; MS: *m/z* 199.12 (M^+).

Ethyl-α-Cyanocinnamate, 6b: Yield 0.171 g (85%). m.p. 51-53ºC; ^{1}H NMR ($CDCl_3$): δ 7.2-8.2 (m, 6H), 4.3 (q, 2H), 1.3 (t, 3H); FT-IR (KBr): 2215.6, 1724.3, 1265.5, 1180.0 cm^{-1}; MS: *m/z* 201.26 (M^+).

(4-Methylbenzylidene)malononitrile, 6c: Yield 0.146 g (87%). m.p. 134-35ºC; ^{1}H NMR ($CDCl_3$): δ 7.9 (s, 1H), 7. (d, 2H), 7.1 (d, 2H),2.3 (t, 3H); FT-IR (KBr): 2218.3, 1595.3 cm^{-1}; MS: *m/z* 168.16 (M^+).

1-Chloro-6-methoxy-2-[β, β-dicyano]-3,4-dihydro-naphthalene, 6d: Yield 0.243 g (90%). m.p. 190-91ºC; ^{1}H NMR ($CDCl_3$): δ 8.3 (s, 1H), 7.4 (d, 1H), 7.1 (d, 1H) 6.5 (s, 1H), 3.8 (s, 3H), 2.6-2.9

(m, 4H); FT-IR (KBr): 2220.1, 1615.0, 1517.3, 1281.4 cm^{-1}; MS: *m/z* 270.72 (M^+).

2-Cyano-5-phenyl-ethylpentadienate, 6e: Yield 0.190 g (84%). m.p. 116-17ºC; ^1H NMR ($CDCl_3$): δ 7.2-7.6 (m, 8H), 4.2 (q, 2H), 1.3 (t, 3H); FT-IR ($CHCl_3$): 2223.0, 1725.1, 1596.7 cm^{-1}; MS: *m/z* 227.19 (M^+).

Cinnamylidenemalononitrile, 6f: Yield 0.153 g (85%). m.p. 126-27ºC; ^1H NMR ($CDCl_3$): δ 7.1-7.6 (m, 8H); FT-IR ($CHCl_3$): 2223.7, 1595.3 cm^{-1}; MS: *m/z* 180.23 (M^+).

(2-Furanylmethylene)malononitrile, 6g: Yield 0.126 g (88%). m.p. 70-74ºC; ^1H NMR ($CDCl_3$): δ 7.2 (s), 7.5(d, 1H), 7.1 (d, 1H), 6.5 (dd, 1H); FT-IR (KBr): 3122.6, 3042.5, 2231.1, 1606.4, 1529.8, 1021.5 cm^{-1}; MS: *m/z* 144.15 (M^+).

α-Cyanocinnamide, 6h: Yield 0.144 g (84%). m.p. 123-24ºC; ^1H NMR ($CDCl_3$): δ 8.1 (s, 1H), 7.1-8.2 (m, 7H); FT-IR (KBr): 3350.6, 2225.8, 1680.2 cm^{-1}; MS: *m/z* 172.22 (M^+).

(4-Bromobenzylidene)malononitrile, 6i: Yield 0.205 g (88%). m.p. 158-61ºC; ^1H NMR ($CDCl_3$): δ 7.4-7.9 (m, 5H); FTIR (KBr): 2226.7, 1578.7 cm^{-1}; MS: *m/z* 233.01 (M^+).

(2,4-dichlorobenzylidene)malononitrile, 6j: Yield 0.187 g (84%). m.p. 144-45ºC; ^1H NMR ($CDCl_3$): δ 7.2-8.0 (m, 4H, Ph); FT-IR (KBr): 2228.4, 1578.8, 1109.9 cm^{-1}; MS: *m/z* 223.09 (M^+).

Acknowledgement

The authors acknowledge the Director, and the Analytical Division of NEIST, Jorhat, Assam, India, for their help. Also, Saikat Das Sharma and Pranjal Gogoi thank the Council of Scientific and Industrial Research (CSIR), New Delhi for the grant of fellowships.

References

1 (a) Li C-J & Chan T-H, *Organic Reactions in Aqueous Media*, (John Wiley and Sons, New York), **1997**; (b) Lindstrom U M, *Chem Rev*, 102, **2002**, 2751; (c) Kobayashi S & Manabe K, *Acc Chem Res*, 35, **2002**, 209.
2 Hara J, Inouye Y & Kakisawa H, *Bull Chem Soc Japan*, 54, **1981**, 3871.
3 Vasella A & Voeffray R, *Helv Chim Acta*, 65, **1982**, 1953.
4 Tufariello J J, Lee G E, Senaratne P A & Al-Nuri M, *Tetrahedron Lett*, **1979**, 4359.
5 DeShong P & Leginus J M, *J Am Chem Soc*, 105, **1983**, 1686.
6 Tufariello J J & Puglis J M, *Tetrahedron Lett*, **1986**, 1489.
7 Huisgen R, *Angew Chem*, 75, **1963**, 604.
8 *1,3-Dipolar Cycloaddition Chemistry*, edited by A Padwa (John Wiley and Sons, Inc, New York), **1984**.
9 Torssell K G B, *Nitrile Oxides, Nitrones, and Nitronates in Organic Synthesis*, (VCH Publishers Inc, New York), **1988**, pp 81-83.
10 a) Doyle M P, Wierenga W & Zaleta M A, *J Org Chem*, 37, **1972**, 1597; (b) Doyle M P, Zaleta M A, Deboer J E & Wierenga W, *J Org Chem*, 38, **1973**, 1663; (c) Ziegler F E, Fowler K W & Kanfer S, *J Am Chem Soc*, 98, **1976**, 8282; (d) Corey E J, Hopkins P B, Kim S, Yoo S, Nambiar K P & Falck J R, *J Am Chem Soc*, 101, **1979**, 7131; (e) Stork G & Dowd S R, *J Am Chem Soc*, 85, **1963**, 2178; (f) Kende A S & Curran D P, *J Am Chem Soc*, 101, **1979**, 1857; (g) Hershberg E B, *J Org Chem*, 13, **1948,** 542.
11 Erickson R E, Andrulis P J Jr, Collins J C, Lungle M L & Mercer G D, *J Org Chem*, 34, **1969**, 2961.
12 Imanzadeh G H, Hajipour A R & Mallakpour S E, *Synth Commun*, 33, **2003**, 735.
13 (a) Rao C G, Radhakrishna A S, Singh B B & Bhatnager S P, *Synthesis*, **1983**, 808; (b) Maloney J R, Lyle R E, Saavedra J E & Lyle G G, *Synthesis*, **1978**, 212; (c) Drabowicz J, *Synthesis*, **1980**, 125; (d) Lee J G, Kwak K H & Hwang J P, *Synth Commun*, 22, **1992**, 2425; (e) Hamal S, Mahto S K & Gajurel C L, *Indian J Chem*, 35 B, **1996**, 1116.
14 Narayanan N & Balasubramaniam T R, *J Chem Res* (S), 4, **1992**, 132.
15 Barton D H R, Lester D J & Ley S V, *J Chem Soc, Chem Commun*, **1977**, 445.
16 Alper H & Amaratunga S, *J Org Chem*, 47, **1982**, 3593.
17 Mori Y, *Micelles Theoretical and Applied*, (Plenum Press, New York and London), **1992**.
18 Manabe K, Limura S, Sun X & Kobayashi S, *J Am Chem Soc*, 124, **2002**, 11971.
19 Kobayashi S & Wakabayashi T, *Tetrahedron Lett*, 39, **1998**, 5389.
20 Chatterjee A, Maiti D K & Bhattacharya P K, *Organic Lett*, 5, **2003**, 3967.
21 (a) Boruah M & Konwar D, *Synlett*, **2001**, 795; (b) Boruah M & Konwar D, *J Org Chem*, 67, **2002**, 7138; (c) Gogoi P, Sarmah G K & Konwar D, *J Org Chem*, 69, **2004**, 5153; (d) Gogoi P, Hazarika P & Konwar D, *J Org Chem*, 70, **2005**, 1934.
22 Konwar D, Boruah R C & Sandhu J S, *Heterocycles*, 23, **1985**, 2557 and references cited therein.
23 Kaminski L S & Lamchen M, *J Chem Soc* (C), **1967**, 1683.
24 Krishnan K & Singh N, *J Indian Chem Soc*, 51, **1974**, 802.
25 *Dictionary of Organic Compounds*, Sixth edn., (Chapman and Hall, London), **1996**.