Synthesis of Some 2-Amino-5-substituted-1,3,4-oxadiazole Derivatives in the Acetic Acid

SANJEEV KUMAR

Department of Chemistry
Isvar Saran Degree College (University of Allahabad)
Allahabad-211004, India
sanjivks77@gmail.com

Received 31 March 2011; Accepted 7 June 2011

Abstract: An environmentally benign method for the preparation of 2-amino-5-substituted-1,3,4-oxadiazoles (**1a-n**) at platinum electrode through the electrooxidation of semicarbazone at room temperature is reported. The electrolysis were carried out at controlled potential in acetic acid solvent using lithium perchlorate as supporting electrolyte in an undivided cell assembly. Square shaped plates of platinum were used for working as well as counter electrode and a third electrode saturated calomel electrode was used as a reference electrode for the measurement of potential difference between two electrodes during the electrolysis.

Keywords: Semicarbazone, 2-Amino-1,3,4-oxadiazoles, Electrooxidation, Controlled potential, Electrolysis

Introduction

2-Amino-5-substituted-1,3,4-oxadiazoles are of considerable pharmaceutical and material interest, which is documented by steadily increasing numbers of publications and patents. 1,3,4-Oxadiazoles have been found to exhibit diverse biological activities such as antibacterial¹⁻³, anti-HIV¹⁻³, anti-microbial¹⁻³, anti-fungal^{4,5}, anti-inflammatory⁶, hypotensive⁷, muscle relaxant⁸ and antimitotic activity⁹.

Methods for the synthesis of oxadiazoles¹⁰⁻¹⁴ (**1**) include bromine oxidation of semicarbazide derivative and the cyclodesulfurisation of acylthiosemicarbazide derivative in solution using I₂/NaOH or 1,3-dicyclohexylcarbodiimide (DCC)¹⁵⁻¹⁹ as well as mercury (II) acetate [Hg(OAc)₂] or yellow mercury (II) oxide HgO²⁰⁻²². All these methods are usually carried out in various different synthetic steps and require heating. The handling of these reagents is not only difficult but also hazardous to the environment. Each stage of the reaction including extraction and purification of the products from the mixture requires great precautions. Evans²³ has synthesized a similar cyclized product by rapid parallel synthesis in efficient one-pot preparation using resin-bound reagents.

Experimental

The purity of the synthesized compounds were ascertained by thin layer chromatography (TLC) on silica gel G 60 F₂₅₄ in various solvent systems using iodine vapor to stain products as detecting agent. All melting points were determined in open capillaries and were uncorrected. Infra red spectra were recorded on a Shimadzu 8201 PC IR spectrophotometer in KBr pellets and reported in cm⁻¹. ¹H NMR and ¹³C NMR spectra were measured at room temperature on Bruker DRX 300 MHz FT spectrometers, with tetramethylsilane (TMS) and CDCl₃ or C₆D₆ as internal standards. Carbon multiplicities were assigned by DEPT techniques. The signals are described as singlet (*s*), doublet (*d*), triplet (*t*), quartet (*q*) and multiplet (*m*). Microanalyses were carried out in the Elementar Vario EL III.

General procedure for the preparation of 2-amino-5-substituted-1,3,4-oxadiazoles (**1a-n**)

Semicarbazide hydrochloride (**3**) (2.0 g, 17.92 mmol) and sodium acetate (1.0 g, 12.19 mmol) was dissolved in 20 mL water. Then aldehyde (**2**) (1.0 g, 6.08 mmol) was added with continuous stirring. The mixture was left overnight, which gave semicarbazone (**4**) as a solid for the electrolysis. Semicarbazone (**4**) (2.0 g, 10.12 mmol) was dissolved in 200 mL acetic acid and lithium perchlorate (0.106 g, 0.76 mmol) was dissolved in the above solution to maintain the strength of supporting electrolyte 0.001 M.

Electrolysis

Preparative scale controlled potential electrolysis²⁴⁻²⁸ was performed at room temperature in 250 mL three-electrode cell assembly with platinum plate as working as well as counter electrode and saturated calomel electrode (SCE) as reference electrode. Magnetic stirrer was used for efficient mixing of reaction mixture. Solid products formed on electrode do not adhere to the electrode surface due to continuous stirring. All the electrolysis experiments were carried out at their corresponding oxidation potentials and were completed in 3 to 5 h. After which no oxidation product was seen to diffuse in the bulk. All the products were solid and coloured and entirely different from the starting compound. The current potential data was recorded with the help of a potentiostat at intervals of 15 min (Table 1). Approximately 4.3×10^5 – 5.8×10^5 C mol⁻¹ of electricity was passed for the electrolysis.

Table 1. Current, potential and yield data for synthesis of 1,3,4-oxadiazoles (**1a-n**)

Entry	R	Compd.	Time, h	Potential, mV	Current, mA	Yield, %
1	4-ClC ₆ H ₄	1a	3	2030	110	82
2	4-FC ₆ H ₄	1b	5	2320	150	86
3	4-(NO ₂)C ₆ H ₄	1c	4	2500	170	85
4	4-(CH ₃ CONH)C ₆ H ₄	1d	3	1350	140	90
5	C ₇ H ₁₅	1e	4	2100	125	87
6	4-Me ₂ NC ₆ H ₄ CHCH	1f	4	1900	150	88
7	(C ₆ H ₅ CO)C ₆ H ₄	1g	5	2000	170	80
8	3-C ₆ H ₅ CO ₂ -(OMe) C ₆ H ₃	1h	5	1750	160	81
9	4-(C ₄ H ₉ O)C ₆ H ₅	1i	3	1800	190	90
10	3,4-(OMe) ₂ C ₆ H ₃	1j	4	1660	110	87
11	4-(AcO)C ₆ H ₄	1k	3	1540	130	95
12	2,4-F ₂ C ₆ H ₃	1l	5	2220	160	77
13	5-Br, 2-(OMe) C ₆ H ₃	1m	5	2100	130	79
14	2-Cl, 6-(NO ₂)C ₆ H ₃	1n	5	2250	170	81

The products were extracted from the acetic acid solution to chloroform by the simple solvent extraction technique. The two-immiscible layers of acetic acid and chloroform were shaken in a separatory funnel and allowed to settle down. After some time the chloroform layer containing the product oxadiazole, was removed. The extracted chloroform layer was separated out with the help of rotatory evaporator and oxadiazole (**1a-n**) were obtained in the excellent yield and purity was checked by the TLC. The product oxadiazole can also be separated out from the reaction solution with the help of column chromatography without using chloroform. This is a more time taking process in comparison to extraction process.

The physical and spectral data of the compounds (**1a-n**)

2-Amino-5-(4-chlorophenyl)-1,3,4-oxadiazole (**1a**)

Brownish crystals, m.p. 81-83 °C, IR (KBr, cm⁻¹) 3360 (NH), 3045 (ArC-H), 1610 (C=N), 1265, 1070 (C-O-C), 600-800 (Ar-Cl), 860, 750 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 7.75 (s, 2H, NH₂), 7.71 (d, 2H, Ar-H₂; ₆, *J* = 8.6 Hz), 7.61 (d, 2H, Ar-H₃; ₅, *J* = 8.6 Hz), ¹³C NMR (DMSO, d₆, δ/ppm): 170.2, 148.5, 141.8, 140.6, 139.5, 129.5, 128.7, 126.5. Anal. calcd. for C₈H₆N₃OCl : C, 49.10; H, 3.06; N, 21.48; Cl, 18.15. Found: C, 48.95; H, 3.00; N, 21.20; Cl, 17.98%.

2-Amino-5-(4-fluorophenyl)-1,3,4-oxadiazole (**1b**)

Brown needles, m.p. 76-78 °C, IR (KBr, cm⁻¹) 3360 (NH), 3055 (ArC-H), 1604 (C=N), 1255, 1080 (C-O-C), 1060 (Ar-F), 950, 860, 740 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 7.75 (s, 2H, NH₂), 7.21 (m, 2H, Ar-H₂; ₆), 7.38 (m, 2H, Ar-H₃; ₅), ¹³C NMR (DMSO, d₆, δ/ppm): 173.1, 163.7 (d, *J*_{C-F} = 250 Hz, ArC₄), 147.7, 147.2, 137.5, 132.3 (d, *J*_{C-F} = 8.6 Hz, ArC₂; ₆), 122.4 (d, *J*_{C-F} = 3.6 Hz, ArC₁), 116.3 (d, *J*_{C-F} = 22 Hz, ArC₃; ₅). Anal. calcd. for C₈H₆N₃OF : C, 53.63; H, 3.35; N, 23.46; F, 10.05. Found: C, 53.25; H, 3.25; N, 23.20; F, 9.95%.

2-Amino-5-(4-nitrophenyl)-1,3,4-oxadiazole (**1c**)

Dark yellow needles, m.p. 81-83 °C, IR (KBr, cm⁻¹) 3341 (NH), 3035 (ArC-H), 1609 (C=N), 1580 (Ar-NO₂), 1272, 1065 (C-O-C), 890, 750, 690 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 7.75 (s, 2H, NH₂), 7.71 (d, 2H, Ar-H₂; ₆, *J* = 8.6 Hz), 7.62 (d, 2H, Ar-H₃; ₅, *J* = 8.6 Hz), ¹³C NMR (DMSO, d₆, δ/ppm): 171.5, 148.1, 147.2, 141.2, 136.4, 134.2, 129.7, 123.7. Anal. calcd. for C₈H₆N₄O₃ : C, 46.60; H, 2.91; N, 27.18. Found: C, 46.10; H, 2.85; N, 26.80%.

2-Amino-5-(4-acetamidophenyl)-1,3,4-oxadiazole (**1d**)

Dark brownish needles, m.p. 78 °C, IR (KBr, cm⁻¹) 3460-3500 (CO-NH₂), 3444 (NH), 2927 (Ar C-H), 2856 (aliphatic C-H), 1611 (C=N), 1270 (C=O), 1250, 1062 (C-O-C), 915, 870, 675 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 8.13 (s, 1H, CONH), 7.75 (s, 1H, NH₂), 7.70 (d, 2H, Ar-H₂; ₆, *J* = 8.6 Hz), 7.61 (d, 2H, Ar-H₃; ₅, *J* = 8.6 Hz), 1.92 (s, 3H, CH₃), ¹³C NMR (DMSO, d₆, δ/ppm): 172.5, 169.5, 147.5, 140.2, 138.9, 128.4, 122.8, 118.6, 118.5, 20.6. Anal. calcd. for C₁₀H₁₀N₄O₂ : C, 55.06; H, 4.58; N, 25.68. Found: C, 54.80; H, 4.43; N, 25.52%.

2-Amino-5-heptyl-1,3,4-oxadiazole (**1e**)

Light yellow crystals, m.p. 83 °C, IR (KBr, cm⁻¹) 3360 (NH), 3060 (C-H), 1608 (C=N), 1280, 1066 (C-O-C), ¹H NMR (DMSO, d₆, δ/ppm): 7.75 (s, 2H, NH₂), 3.5 (t, 2H, CH₂ (CH₂)₅CH₃, *J* = 7.3 Hz), 1.5 (sextet, 2H, CH₂(CH₂)₄CH₂CH₃, *J* = 7.3 Hz), 1.2-1.8 (quintet, 8H, CH₂ (CH₂)₄CH₂CH₃, *J* = 7.3 Hz), 0.9 (t, 3H, CH₂ (CH₂)₅CH₃, *J* = 7.3 Hz), ¹³C NMR (DMSO, d₆, δ/ppm): 170.8, 148.6, 147.6, 32.2, 32.1, 23.5, 23.3, 23.1, 14.1. Anal. calcd. for C₉H₁₇N₃O : C, 59.01; H, 9.28; N, 22.95. Found: C, 58.76; H, 9.15; N, 22.60%.

2-Amino-5-(4-*N,N*-dimethylaminocinnamyl)-1,3,4-oxadiazole (If)

Dark blackish crystals, m.p. 71-73 °C, IR (KBr, cm⁻¹) 3336 (NH), 3033 (=C-H), 3030 (ArC-H), 2855 (aliphatic C-H), 1670 (C=C), 1606 (C=N), 1262, 1067 (C-O-C), 920, 860, 790 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 7.75 (s, 2H, NH₂), 7.71 (d, 2H, Ar-H_{2,6}, *J* = 8.6 Hz), 7.61 (d, 2H, Ar-H_{3,5}, *J* = 8.6 Hz), 6.52 (d, 1H, PhCH=CH, *J* = 16.8 Hz), 6.39 (d, 1H, PhCH=CH, *J* = 16.8 Hz), 2.35 (s, 6H, CH₃), ¹³C NMR (DMSO, d₆, δ/ppm): 170.6, 147.3, 146.5, 138.1, 137.6, 129.2, 129.1, 126.9, 126.5, 14.2, 13.7. Anal. calcd. for C₁₂H₁₄N₄O : C, 62.60; H, 6.08; N, 24.34. Found: C, 62.50; H, 5.90; N, 24.25%.

2-Amino-5-(2-benzoylphenyl)-1,3,4-oxadiazole (Ig)

Brownish needles, m.p. 70-72 °C, IR (KBr, cm⁻¹) 3350 (NH), 3045 (ArC-H), 1755 (C=O), 1620 (C=N), 1273, 1059 (C-O-C), 880, 820, 785 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 7.98 (d, 2H, Ar-H, *J* = 8.6 Hz), 6.93-7.03 (m, 2H, Ar-H), 7.75 (s, 2H, NH₂), 7.45-7.56 (m, 3H, Ar-H), 7.38-7.42 (m, 2H, Ar-H), ¹³C NMR (DMSO, d₆, δ/ppm): 171.2, 168.6, 149.5, 137.1, 136.3, 132.9, 131.3, 130.2, 128.7, 128.5, 128.1, 128.0, 127.8. Anal. calcd. for C₁₅H₁₁N₃O₂ : C, 67.92; H, 4.15; N, 15.84. Found: C, 67.60; H, 4.10; N, 15.65%.

2-Amino-5-(3-benzoyl-2-methoxyphenyl)-1,3,4-oxadiazole (Ih)

Brown crystals, m.p. 75-76 °C, IR (KBr, cm⁻¹) 3261 (NH), 3045 (ArC-H), 2815 (OCH₃), 1760 (C=O), 1609 (C=N), 1270, 1069 (C-O-C), 915, 870, 790 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 8.75 (s, 1H, Ar-H), 7.98 (d, 2H, Ar-H, *J* = 8.6 Hz), 7.75 (s, 2H, NH₂), 7.45-7.56 (m, 3H, Ar-H), 6.95-7.41 (m, 3H, Ar-H), 6.93-7.03 (m, 3H, Ar-H), 3.11 (s, 3H, OCH₃), ¹³C NMR (DMSO, d₆, δ/ppm): 172.6, 168.7, 147.5, 137.1, 136.3, 133.1, 132.9, 131.3, 129.5, 128.5, 128.2, 128.0, 127.8, 114.6, 52.3. Anal. calcd. for C₁₆H₁₃N₃O₃ : C, 65.08; H, 4.90; N, 14.23. Found: C, 64.88; H, 4.57; N, 14.15%.

2-Amino-5-(4-butoxyphenyl)-1,3,4-oxadiazole (Ii)

Yellow crystals, m.p. 69-71 °C, IR (KBr, cm⁻¹) 3261 (NH), 3060 (C-H), 2820 (OC₄H₉), 1608 (C=N), 1269, 1067 (C-O-C), 950, 890, 770 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 7.75 (s, 2H, NH₂), 7.71 (d, 2H, Ar-H_{2,6}, *J* = 8.6 Hz), 7.63 (d, 2H, Ar-H_{3,5}, *J* = 8.6 Hz), 3.8 (t, 2H, CH₂(CH₂)₂CH₃, *J* = 7.3 Hz), 2.3 (Quintet, 2H, CH₂CH₂CH₂CH₃, *J* = 7.3 Hz), 1.6 (sextet, 2H, CH₂CH₂CH₂CH₃, *J* = 7.3 Hz), 0.9 (t, 3H, CH₂CH₂CH₂CH₃, *J* = 7.3 Hz), ¹³C NMR (DMSO, d₆, δ/ppm): 172.3, 149.9, 142.8, 129.5, 128.6, 127.6, 114.2, 114.1, 65.6, 54.5, 23.8, 10.0. Anal. calcd. for C₁₂H₁₅N₃O₂ : C, 61.80; H, 6.43; N, 18.02. Found: C, 61.45; H, 6.23; N, 17.95%.

2-amino-5-(3,4-dimethoxyphenyl)-1,3,4-oxadiazole (Ij)

Dark brownish needles, m.p. 77-79 °C, IR (KBr, cm⁻¹) 3444 (NH), 2927 (Ar C-H), 2856 (aliphatic C-H), 2865 (O-CH₃), 1605 (C=N), 1268, 1074 (C-O-C), 895, 860, 790 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 8.76 (s, 1H, Ar-H), 7.91 (d, 1H, Ar-H, *J* = 8.2 Hz), 7.75 (s, 2H, NH₂), 7.30 (d, 1H, Ar-H, *J* = 8.2 Hz), 3.10-3.73 (s, 6H, OCH₃), ¹³C NMR (DMSO, d₆, δ/ppm): 171.7, 148.8, 143.9, 143.4, 136.6, 133.7, 121.8, 116.5, 54.1, 46.7. Anal. calcd. for C₁₀H₁₁N₃O₃ : C, 54.29; H, 4.97; N, 19.00. Found: C, 54.00; H, 4.73; N, 18.75%.

2-amino-5-(4-acetoxyphenyl)-1,3,4-oxadiazole (Ik)

Light brown crystals, m.p. 78-79 °C, IR (KBr, cm⁻¹) 3330 (NH), 3045 (ArC-H), 2855 (aliphatic C-H), 1720 (C=O), 1607 (C=N), 1285 (C-O), 1265, 1069 (C-O-C), 910, 860, 735 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 7.75 (s, 2H, NH₂), 7.72 (d, 2H, Ar-H_{2,6}, *J* = 8.6 Hz),

7.63 (d, 2H, Ar-H_{3,5}, $J = 8.6$ Hz), 1.48 (s, 3H, CH₃), ¹³C NMR (DMSO, d₆, δ/ppm): 172.2, 165.3, 146.5, 137.8, 136.5, 128.8, 127.8, 126.5, 54.5, 20.6. Anal. calcd. for C₁₀H₉N₃O₃: C, 54.79; H, 4.10; N, 19.17. Found: C, 53.94; H, 4.05; N, 19.10%.

2-Amino-5-(2,4-difluorophenyl)-1,3,4-oxadiazole (**II**)

Dark brownish crystals, m.p. 80-82 °C, IR (KBr, cm⁻¹) 3360 (NH), 3050 (ArC-H), 1615 (C=N), 1259, 1068 (C-O-C), 1100-1400 (Ar-F), 940, 870, 770 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 8.77 (s, 1H, Ar-H), 7.75 (s, 2H, NH₂), 7.15-7.47 (m, 2H, Ar-H), ¹³C NMR (DMSO, d₆, δ/ppm): 171.8, 164.9, 149.5, 142.5, 139.4, 125.6, 122.2, 116.5, 100.1. Anal. calcd. for C₈H₅N₃O₂F₂: C, 48.73; H, 2.53; N, 21.31; F, 19.28. Found: C, 48.67; H, 2.40; N, 21.10; F, 19.05%.

2-amino-5-(5-bromo-2-methoxyphenyl)-1,3,4-oxadiazole (**Im**)

Brownish crystals, m.p. 75-77 °C, IR (KBr, cm⁻¹) 3360 (NH), 3045 (ArC-H), 2860 (aliphatic C-H), 2820 (O-CH₃), 1613 (C=N), 1265, 1072 (C-O-C), 980, 890, 750, 595 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 8.78 (s, 1H, Ar-H), 7.89 (d, 1H, Ar-H, $J = 8.2$ Hz), 7.75 (s, 2H, NH₂), 7.28 (d, 1H, Ar-H, $J = 8.2$ Hz), 3.11 (s, 3H, OCH₃), ¹³C NMR (DMSO, d₆, δ/ppm): 172.2, 158.9, 147.7, 140.6, 132.9, 125.1, 115.4, 106.4, 53.8. Anal. calcd. for C₉H₈N₃O₂Br: C, 40.00; H, 2.96; N, 15.55; Br, 29.62. Found: C, 39.52; H, 2.46; N, 15.30; Br, 29.56%.

2-Amino-5-(2-chloro-6-nitrophenyl)-1,3,4-oxadiazole (**In**)

Dark yellowish needles, m.p. 71-73 °C, IR (KBr, cm⁻¹) 3341 (NH), 3035 (ArC-H), 1607 (C=N), 1580 (Ar-NO₂), 1275, 1070 (C-O-C), 985, 865, 810, 730 (substituted benzene), ¹H NMR (DMSO, d₆, δ/ppm): 7.75 (s, 2H, NH₂), 7.05-7.19 (m, 3H, Ar-H), ¹³C NMR (DMSO, d₆, δ/ppm): 171.6, 148.3, 147.1, 146.8, 140.8, 136.8, 130.4, 121.2. Anal. calcd. for C₈H₅N₄O₃Cl: C, 39.91; H, 2.07; N, 23.28; Cl, 14.76. Found: C, 39.60; H, 2.00; N, 23.15; Cl, 14.50%.

Results and Discussion

An environmentally benign synthetic method to produce 2-amino-5-substituted-1,3,4-oxadiazoles would be highly attractive. We have synthesized 2-amino-5-substituted-1,3,4-oxadiazole (**1**) derivatives through the electrooxidation of semicarbazone (**4**) at a platinum electrode in controlled potential electrolysis. The reaction sequences employed for the synthesis of title compound is shown in the Scheme 1. This electrochemical cyclization gives the oxadiazoles without requirement of any hazardous reagents. We have used acetic acid as a solvent and lithium perchlorate (LiClO₄) (0.001 M) as an electrolyte that can be handled very easily without major precautions.

Scheme 1

Scheme 2

However, to induce such cyclizations electrolytically has various merits. The electrochemical oxidation reactions do not require oxidizing chemicals because the oxidation takes place at the electrode and, furthermore, can be performed under mild conditions, *e.g.*, at room temperature. Application of electricity as a non conventional energy source for activation of reactants in suitable solvents has now gained popularity over the usual homogeneous and heterogeneous reactions. It provides chemical processes with special attributes, such as enhanced reaction rate, higher yield of pure products, better selectivity and several ecofriendly advantages.

Mechanistic proposals

The first step as shown in Scheme 2 represents the deprotonation of (4) to form an anion (4a), which rearranges itself into (4b) and evolves a free radical (4c) after one electron oxidation. Subsequent second electron oxidation from the free radical (4c) gives a carbocation (4d). The formation of carbon oxygen bond completes the ring. On losing a proton in the last step 2-amino-5-substituted-1,3,4-oxadiazole (1) is obtained.

2-Amino-5-substituted-1,3,4-oxadiazoles have been synthesized in excellent yields using the synthetic route outlined in Scheme 1. IR, ^1H NMR, ^{13}C NMR and mass spectral data are in good agreement with the proposed structures of all the synthesized compounds. Lack of ^1H NMR resonances observed with NH and NH_2 functions in the ^1H NMR spectrum of (1a-n) proved that ring closure starting from (4) resulted in the formation of 1,3,4-oxadiazole ring. This was further substantiated by the ^{13}C NMR data of (1) which showed the peaks at δ 170-173 and 145-150 due to C_2 and C_5 of oxadiazole respectively. Infra red spectrum shows $1600\text{--}1620\text{ cm}^{-1}$ for ($\text{C}=\text{N}-\text{N}=\text{C}$) and $1063\text{--}1073\text{ cm}^{-1}$ for ($\text{C}-\text{O}-\text{C}$) in the compounds (1a-n) which confirmed the synthesis of 1,3,4-oxadiazoles.

Conclusion

In conclusion we have developed a convenient method for the synthesis of novel 2-amino-5-substituted-1,3,4-oxadiazole derivatives in excellent yields which is not so easy to achieve by

chemical methods. In the present electrolytic method, electrolysis was carried out at ordinary temperature and no hazardous chemicals were used. Therefore the method is environmentally benign and the great contribution in the field of green chemistry.

Acknowledgment

The author gratefully acknowledge the financial assistance provided by University Grant Commission, New Delhi and to the Sophisticated Analytical Instrumentation Facility (SAIF), A division of CDRI (Central Drug Research Institute) Lucknow, for providing spectra and microanalyses.

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