This article was downloaded by: [UZH Hauptbibliothek / Zentralbibliothek Zürich] On: 29 December 2014, At: 03:27 Publisher: Taylor & Francis Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



Phosphorus, Sulfur, and Silicon and the Related Elements

Publication details, including instructions for authors and subscription information:

http://www.tandfonline.com/loi/gpss20

IMPROVED ONE-POT SYNTHESIS OF 3-SPIRO INDOLINES UNDER MICROWAVE IRRADIATION

Anshu Dandia^a, Mitali Saha^a & Harshita Taneja^a

 $^{\rm a}$ Department of Chemistry , University of Rajasthan , Jaipur, 302004, India

Published online: 16 Feb 2007.

To cite this article: Anshu Dandia , Mitali Saha & Harshita Taneja (1998) IMPROVED ONE-POT SYNTHESIS OF 3-SPIRO INDOLINES UNDER MICROWAVE IRRADIATION, Phosphorus, Sulfur, and Silicon and the Related Elements, 139:1, 77-85, DOI: <u>10.1080/10426509808035679</u>

To link to this article: <u>http://dx.doi.org/10.1080/10426509808035679</u>

PLEASE SCROLL DOWN FOR ARTICLE

Taylor & Francis makes every effort to ensure the accuracy of all the information (the "Content") contained in the publications on our platform. However, Taylor & Francis, our agents, and our licensors make no representations or warranties whatsoever as to the accuracy, completeness, or suitability for any purpose of the Content. Any opinions and views expressed in this publication are the opinions and views of the authors, and are not the views of or endorsed by Taylor & Francis. The accuracy of the Content should not be relied upon and should be independently verified with primary sources of information. Taylor and Francis shall not be liable for any losses, actions, claims, proceedings, demands, costs, expenses, damages, and other liabilities whatsoever or howsoever caused arising directly or indirectly in connection with, in relation to or arising out of the use of the Content.

This article may be used for research, teaching, and private study purposes. Any substantial or systematic reproduction, redistribution, reselling, loan, sub-licensing, systematic supply, or distribution in any form to anyone is expressly forbidden. Terms & Conditions of access and use can be found at http://www.tandfonline.com/page/terms-and-conditions

Phosphorus, Sulfur and Silicon, 1998, Vol. 139, pp. 77-85 Reprints available directly from the publisher Photocopying permitted by license only

IMPROVED ONE-POT SYNTHESIS OF 3-SPIRO INDOLINES UNDER MICROWAVE IRRADIATION

ANSHU DANDIA^{*}, MITALI SAHA and HARSHITA TANEJA

Department of Chemistry, University of Rajasthan, Jaipur 302004, India

(Received 15 September, 1997; In final form 24 November, 1997)

The potential of the domestic microwave oven has been utilized to accelerate the one-pot synthesis of spiro[indole-thiazolidine]diones and spiro[indole-benzothiazine]diones by condensation of substituted indole-2,3-diones (1) and appropriate anilines (2) with mercaptopropionic acid (4a/b) and o-mercaptobenzoic acid (5) in open borosilicate vessels, using ethanol as energy transfer medium. The reaction rate was 400–500 times faster than the reaction rate in the conventional way. Excellent isolated yields with easier workup than classical heating were the main advantages observed.

Keywords: Microwaves; indole-2,3diones; spiro[indole-thiazolidine]; spiro[indolebenzothi-azine]

INTRODUCTION

The great importance of the indole nucleus in the field of medicinal chemistry has attracted the attention of chemists for a long time.¹⁻³ Indole derivatives have acquired conspicuous popularity in recent years due to their wide spectrum of biological activities⁴ including anti-convulsant and anti-inflammatory activities. Literature surveys reveal a wide range of pharmacological properties affiliated with compounds in which an indole ring is joined with another heterocycle at the C-3 position through a spiro carbon atom.^{5–8}

Several naturally occurring alkaloids, e.g. elegantine, rhynchophylline and surgatoxin, are heterocyclic compounds with a spiro atom at position 3

^{*} To whom correspondence should be addressed.

ANSHU DANDIA et al.

of the 2-indoline skeleton. Fredricamycin-A, a spiro heterocycle, is an antitumor antibiotic agent. Strychofline, isolated from Strychnos usambarensis has been found to exhibit antimitotic activity of cancer cell culture.⁹

Diverse types of biological activity are displayed by benzothiazines¹⁰ and thiazolidinones.¹¹ The presence of the -N-C-S linkage has been postulated to account for the antifungal activity of 4-Thiazolidinones¹². The 2-pyridyl thiazolidin-4-one derivatives act as antiulcer agents¹³. Spiro [indole-thiazolidines] were found to possess a wide variety of biological activity.^{5-8,14-16} These compounds have been synthesized earlier by a two step procedure in 40-60% yield.¹⁷⁻²¹. Popp²² et al. have also synthesized a number of 5'-methyl-spiro [3H-indole-3,2'-thiazolidine]-2,4' (IH)-diones in order to investigate their potential as anticonvulsants. The general synthetic approach involved the preparation of isatin-3-imines from substituted isatins and a number of aromatic amines. They were then subjected to cyclocondensation with thiolactic acid in toluene under reflux for 10 hours, with azeotropical removal of the water formed, to give the desired spiro compounds in 26–60% yields. Joshi²³ et al. reported the reactions of 3-arylimino-2H-indol-2-ones with o-mercaptobenzoic acid in refluxing alcohol-acetic acid (50: 10) for 10 hours to give 4,5-dihydro-3-phenyl spiro-[2H-1,3-benzothiazine-2,3'-[3Hj-indole]-2',4 (1'H,3H-diones in 70-78% yield.

Application of microwave irradiation to improve organic syntheses, is of recent origin^{24,25}, leading to shorter reaction times, higher yields, cleaner reaction products and easier work-up than classical heating²⁶. Microwave heating has been used for a wider variety of purposes²⁷ such as moisture analysis, the wetashing of biological and dissolution of geological methods, regeneration of activated carbon, the preparation of activated carbon from carbonaceous materials, the treatment of sewage and sewage sludge etc, but the opportunities which such technology offers to synthetic organic chemists, were first highlighted in two important contribution from Gedye, Smith and Westaway²⁸ and Giguere, Majetich and coworkers²⁹. They demonstrated that many organic reaction can be conducted very rapidly under microwave irradiation using a commercially available microwave oven which are now widely accepted as safe and convenient devices in the kitchen.

Most of the reactions carried out in an unmodified domestic microwave oven are either in sealed vessels³⁰ or in the solid phase³¹. This method employs polar, high-boiling solvents in open vessels, using organic sol-

3- SPIRO INDOLINES

vents like ethanol, N,N-dimethyl-formamide (DMF), 1,2-dichloroethane (DCE), o-dichlorobenzene etc. as energy transfer media. Usually a solvent is chosen with dielectric properties suitable for efficient coupling of microwave energy, and rapid heating to temperatures, which although high, are typically some $20-30^{\circ}$ C below boiling.³²

As an extension of our research work on the synthesis of biologically active spiro indoles^{33–37} and earlier interest on the cyclocondensation reaction of arylimines with thio acids^{34,38–41}, we wish to report herein, an improved one-pot elegant synthesis of 3'-phenyl spiro [3H-indole-3,2'-thi-azolidine]-2,4' (1H)-diones and 4,5-dihydro-3-phenyl-spiro [2H-1,3-ben-zothiazine-2,3'-[3H]-indole]-2',4-(1'H,3H)-diones under microwave irradiation. It may be mentioned here that this is the first report on the synthesis of spiro heterocycles, where the microwave technique has been used, and the results demonstrate the versatility of the process with faster reactions, easy workup and enhanced yields.

RESULTS AND DISCUSSION

Various 3'-phenyl spiro [3H-indole-3,2'-thiazolidine]-2,4' (1H)-diones (6) 4,5-dihydro-3-phenyl and two spiro[2H-1 .3-benzothiazine-2,3'-[3H]-indole]-2',4 (1'H,3H)-diones (7) were synthesized in one-step under microwave using either ethanol as the energy transfer medium or adsorbing the reactants on silica gel as inorganic solid support. The condensation of appropriate indole-2,3-diones (1) with anilines (2) yielded, 3-aryl imino-2H-indol-2-ones (3) which, in situ, were cyclized with mercaptoacetic acid (4 a) /2-mercapto propionic acid (4 b) to give the spiro compounds (6) in 54-87% yields, while a parallel reaction of suitably substituted isatins and anilines with o-mercaptobenzoic acid (5), afforded (7) in 82-89% yields.

A comparison of the time required for the reaction reported in (Table I) clearly indicates a tremendous reduction in the time of reaction in the microwave oven. The efficiency of these condensation reactions was due to the good-dielectric coupling of polar 3-indolylimines and water formed during the reactions, which was vaporized leading to spiro indolines with a bad dielectric coupling. The condensation was adjusted under microwave by the specific dielectric coupling. Moreover, since the polar molecules are immediately vaporized this obviates the use of a Dean-Stark apparatus.

ANSHU DANDIA et al.

S. No.	X	R ¹	R	Reaction time			Lit.	Yield (%)	
				MW (min.)	Standard (hrs.)	(°C)	m.p. ^{22,23} (°C)	MW	Std.
6a	Н	CH ₃	Н	6	9	200-202	200-201	72	57
6b	Н	CH ₃	4-OCH ₃	9	10	180–182	183-184	69	58
6c	Н	CH ₃	4-C1	7	9	245246	246–247	70	53
6d	5-NO ₂	CH ₃	4-Cl	10	11	276–278(d)	277(d)	54	38
6e	5-CH ₃	CH ₃	4-Cl	7	10	251-252	252	78	54
6f	Н	Н	4-F	8	9	242-244	244	87	75
6g	5-Cl	н	3-Cl	6	4	134-136	136	83	72
7a	н	-	н	8	10	285-287	286	89	78
7b	Н	-	4-CH ₃	7	10	279-280	280	82	72

TABLE I Results of synthesis of (6) and (7) under microwave

d = decomposes.

In order to study the stereoeffects, the substituents were varied on both side of the indole and aryl ring, as their attachment may facilitate or retard the condensation through electronic effects. It was observed that bulky and electron with drawing groups on either side hinder the condensation and required more time for completion of the reaction.

EXPERIMENTAL

Infra red spectra were recorded on Perkin Elmer (Model-577) in KBr pellets. ¹H and ¹⁹FNMR were recorded on Jeol (model-FX 90 Q) using CDCl₃as solvent at 89.55 and 84.25 MHz respectively. TMS was used as internal reference for ¹HNMR and hexa fluorobenzene as external reference for ¹⁹FNMR. Mass spectra were recorded on kratos 30 and 50 mass spectrometers. Melting point (Mp) in °C are uncorrected. All compounds were found homogeneous on TLC in various solvent systems.

3'-Phenylspiro [3H-indole-3,2'-thiazolidinel-2,4' (1H)-diones (6a-g)

6a has been synthesized by three different methods under microwave irradiation. The percentage yields and reaction time varied in each case.

Method A

The compound has been synthesized in two step under microwave irradiation, following the method of Popp et al^{22}

(a) Synthesis of 3-arylimino-2H-indol-2-one(3a)

A mixture of indole-2,3-dione 1a (0.01 mole) and aniline 2 (0.01 mole) in absolute ethanol (20 ml) taken in a borosil beaker (250 ml) was zapped inside a microwave oven at 360 watts, i.e. 30% power for 3 minute. On cooling, crystals separated out which was filtered, dried and recrystallized from ethanol, m.p. 142°C; yield 74%.

(b) Synthesis of 5'-'-phenl spiro[3H-indole-3, 2'-thiazolidine]-2,
4'(1H)-dione (6a)

The mixture of 3a (0.01 mole) and 4b (0.015 mole) in absolute ethanol (20 ml) was irradiated at 480 watts, i.e. 40% power for 2 min. After heating, an interval of 30s is allowed in order to avoid excessive evaporation of solvent. Then the heating was continued for 5 min. The progress of the reaction was monitored by TLC. The reaction mixture was cooled and the product separated was filtered, dried and recrystallized from ethanol, m.p. 200°c, yield 63%; lit²² m.p. 200-201, yield 57%.

Method B

The compound has been synthesized in one step, without isolation of anil-

A mixture of 1a (0.01 mole) and 2 (0.01 mole) in absolute ethanol (20 ml) was irradiated in open borosil beaker (250 ml) at (360 watts, 30% power) for 3 min. The progress of the reaction was monitored by TLC. As the reactants disappear, 4b (0.015 mole) was added and then again irradiated at (480 watts, 40% power). After every 2 min. an interval of 30s is allowed, to avoid excessive evaporation of solvent and the heating was continued up to 6 min. The mixture was then cooled, the solid separated was filtered and recrystallized from ethanol to give 6a, m.p. 200°C; yield 72%.

Method C

(a) Synthesis of anil (3a) in solid phase

The equimolar mixture of la (0.01 mole) and 2 (0.01 mole) was adsorbed onto neutral alumina (5 gm). The well powdered and spread reaction mixture was irradiated at (600 watts, 50% power) for 9 min. Reaction was

ANSHU DANDIA et al.

monitored by TLC. The mixture was dissolved in ethanol and the filtrate on concentrated gave the anil 3a, m.p. 142°C; yield 72%.

(b) Synthesis of spiro compound in solid phase

When the mixture of isatin-3-anil 3a (0.01 mole) and 4b (0.015 mole) was adsorbed onto solid support silica gel and irradiated at (480 watts, 40% power), a sticky solid compound was obtained after 7 min. which was treated with petroleum-ether and filtered. The filtrate on concentration gave a solid compound which was purified by recrystallization from ethanol, m.p. 200°C; yield 69%.

The indentity of the product was established by their mixed melting points, co-IR and co-NMR. The remaining compounds were synthesized by method B only, since it involved easy work-up and better yield of desired compounds.

4,5-Dihydro-3-phenyl spiro[2H-1,3-benzothiazine-2,3'-[3H]-indole]-2',4 (l'H,3H)-diones (7a-b)

The synthesis of the compound 7a was tried by following two methods.

Method A

The compound has been synthesized in two steps under microwave irradiation, following the method of Joshi et al^{23} .

(a) The 3-arylimino-2H-indole-2-one 3a was synthesized under microwaves as described earlier.

(b) Synthesis of 4,5-dihydro-3-phenyl spiro [2H-1,3-benzothiazine-2,3'-[3H]-indole]-2[4(1'H,3H)-drones (7a)

The mixture of 3a (0.01 mole) and o-mercaptobenzoic acid 5 (0.01 mole) in absolute ethanol (25 ml) with 2–3 drops of glacial acetic acid, was irradiated at 480 watts for 6 min. The reaction mixture was cooled and the solid separated was filtered, dried and recrystallized from ethanol, m.p. 285°C, yield 82%; Lif²³ m.p. 286, yield 78%.

Method B

7a has also been synthesized in one step, without isolation of anil 3a.

3- SPIRO INDOLINES

A mixture of la (0.01 mole) and 2 (0.01 mole) in absolute ethanol (20 ml) was zapped inside a microwave oven at 360 watts, 30 % power for 3 min, and the reaction was monitored by TLC. As the reactants disappear, 5 (0.01 mole) and 2–3 drops of glacial acetic acid were added and again irradiated at (480 watts, 40% power) for 5 min., with short interruption of 30s. On cooling, a solid compound was separated, which was filtered, dried and recrystallized from ethanol, m.p. 285°C, yield 89%.



7b was prepared by method B only.

Compound 6 a : $C_{17}H_{14}N_2O_2S$: ¹HNMR (CDCl₃) : δ 2.1–2.26 (d, 3H, CH-CH₃), 4.61–4.82 (q, 1H, CH-CH₃), 6.7–7.54 (m, Ar-H), 9.07 (S, 1H, NH).

Compound 6 b : $C_{18}H_{13}N_2O_3S$: ¹HNMR (CDCl₃) : δ 2.1–2.20 (d, 3H, CH-CH₃), 4.5–4.61 (q, 1H, CH-CH₃), 3.85 (S, 3H, OCH₃), 6.8–7.55 (m, Ar-H), 9.09 (S, 1H, NH).

Compound 6 c : $C_{17}H_{13}N_2O_2SCl$: ¹HNMR (CDCl₃) : δ 2.0–2.2 (d, 3H, CH-CH₃), 4.62–4.84 (q, 1H, CH-CH₃), 6.75–7.54 (m, Ar-H), 9.02 (S, 1H, NH).

Compound 6 d : $C_{17}H_{12}N_3O_4SCI$: ¹HNMR (CDCl₃) : δ 2.2–2.35 (d, 3H, CH-CH₃), 4.5–4.79 (q, 1H, CH-CH₃), 6.8–7.0 (m, Ar-H), 9.1 (S, 1H, NH).

Compound 6 e : $C_{18}H_{15}N_2O_2SCI$: ¹HNMR (CDCl₃) : δ 1.1 (S, 3H, CH₃), 2.1–2.3 (d, 3H, CH-CH₃), 4.62–4.83 (q, 1H, CH-CH₃), 6.7–7.1 (m, Ar-H), 9.04 (S, 1H, NH).

Compound 6 f: $C_{16}H_{11}FN_2O_2S$: ¹HNMR(CDCl₃): 64.02 (dd, 2H, CH₂), 6.8–7.4 (m,Ar-H), 9.012 (s,1H,NH); MS m/z (%) : 314 (M+;24) 286 (38) 271 (46.5) 219 (57) 132 (96) 104 (92) 89 (56).

Compound 6 g : $C_{16}H_{10}N_2O_2Cl_2$: ¹HNMR (CDCl₃) : δ 4.03 (dd, 2H, CH₂), 6.9–7.9 (m, Ar-H), 9.05 (S, 1H, NH).

Compound 7 a: $C_{21}H_{14}N_2O_2S$: ¹HNMR (CDCl₃): δ 7.17–8,1 (m, Ar-H), 10.0 (S, 1H, NH).

Compound 7 b : $C_{22}H_{16}N_2O_2S$: ¹HNMR (CDCl₃) : 6 7.18–8.24 (m, Ar-H), 10.05 (S, 1H, NH).

Acknowledgements

Financial assistance from U.G.C., New Delhi is gratefully acknowledged.

References

- a) S. Kakimoto and J. Nishie, Japan, J. Tuberc, 2, 334 (1954); b) V.M. Sim., J. Forensic Sci., 6, 31 (1961); c) C.A. Demerson, L.C. Humber, A.H. Phillip and R.P. Martel, J. Med Chem., 19, 391 (1976); d)H.M. Deubel, D.G.G. Wolk, Einstein and H.H. Von Dobeneek, Brit. Pat., 1313675 (1973); Chem. Abstr, 79, 42357 (1973).
- [2] a) M.E. Flaugh, U.S. Pat., 4614807 (1987); Chem. Abstr., 106, 4768 (1987); b) Y.C. Cong, and K.F. Cheng, U.K. Pat., 2207670 (1989); Chem. Abstr., 111, 115662 (1989).
- [3] a) H.J. Bein, *Pharmacol Rev.*, 8, 435 (1967); b) A. Hofmann and A. Berger, "Drugs Affecting the Central Nervous System," Ed. Dekker, New York, 1968.
- [4] R. Agarwal, C. Agarwal, C. Singh and V.S. Mishra, Indian J. Chem., 28 B, 893 (1989).
- [5] M. Wolf and A.A. Mascitti, U.S. Pat., 3, 458 (1969); Chem. Abstr., 72, 21715 p (1970).
- [6] G.C. Rovnyak, V.L. Narayanan and R.D. Haugwitz, U.S. Pat., 4, 053, 613 (1977); Chem. Abstr., 88, 22889 r (1978).
- [7] B.M. Kirichenko, A.V. Vhadzimirskaya and P.M. Steblyuk, Fann. Zh. (Kiev). 61(1981), Chem. Abstr., 95, 169060 (1981).
- [8] B.J. Sweetman, M. Bellas and L. Field, J. Medhl. Chem., 12, 888 (1969).
- [9] R.P. Bassleer, M. Caprasser and L. Angenal, Planta Med., 45 (2), 123 (1982)
- [10] P.P. Llobet, E. Baylina, Chem. Abstr., 106, 18582 (1986); K., Kikazawa, H. Hiiragi, O. Intro, K. Nakazato, S. Kanezuka, S. Oba, K. Wakizzka, Y. Marayama and M. Riyutsu, Chem. Abstr., 106, 18583 (1986); J.B. Orpi, T. Iso and M. Oya, Chem. Abstr., 102, 24637(1985); G. Benz, G. Fengler, H. Meyer, E. Niemers, V Fiedler, M. Mardin, D. Mayer, E. Perzborn and F. Senter, Chem. Abstr., 105, 60620 (1986).
- [11] J.N. Hriband and J.G. Jurcak, Eur. Pat. Appl., Ep 316, 72324 (1989); Chem. Abstr., 112, 35888 d (1990).
- [12] K.P. Jadhav and D.B. Ingle, J. Indian Chem. Soc., 55, 424 (1978).
- [13] N. Masai, M. Enomotu, A. Kojima, H. Masumori, N. Hara, Y Hara and S. Monooka, Eur. Pat. Appl. Ep., 325, 496 (1989); Chem. Abstr., 113, 1152954 (1990).

3- SPIRO INDOLINES

- [14] F.D. Popp and M. Rajopadhye, J. Heterocycl Chem., 21, 289 (1984).
- [15] E. Piscopo, M.V. Diurno, O. Mazzoni, A.M. Ciaccio and G. Veneruso, Boll-Soc. Ital. Biol. Sper., 66 (12), 1187 (1990); Chem. Abstr., 116, 169945 m (1992).
- [16] E. Piscopo, M.V. Diurno, O. Mazzoni and A.M.Ciaccio, Boll.-Soc. Ital. Biol. Sper., 66 (12), 1181 (1990); Chem. Abstr., 116, 169944 k (1992).
- [17] H. Otomasu and S. Ohmlya, Jap. Pat., 75, 140, 441 (1975); Chem. Abstr., 85, 32991 v (1976).
- [18] K.M. Hassan, A.K. El-Shafei and H.S. El-Kaehef, Z. Naturforsch., B. Anorg. Chem., Org. Chem., 33 B, 1508 (1978); Chem. Abstr., 90, 121763 t (1979).
- [19] K.M. Hassan, A.K. El-Shafei and H.S. El-Kaehef, *ibid.*, **33 B**, 1515 (1978); *Chem. Abstr.*, **90**, 121474 (1979).
- [20] K.M. Hassan and Z.H. Khalil, *ibid.*, **34 B**, 621 (1979); *Chem. Abstr.*, **91**, 56891 n (1979).
- [21] K.M. Hassan and Z.H. Khalil *ibid.*, **34 B**, 1326 (1979); Chem. Abstr., **92**, 146666 t (1980).
- [22] M. Rajopadhye and F.D. Popp., J. Hetemcyclic Chem., 21, 289 (1984).
- [23] K.C. Joshi, R. Jain and S. Nishith, J. Indian, Chem. Soc., 68, 625 (1991).
- [24] D.M.P. Mingos and D.R. Baghurst; Chem. Soc. Rev., 20, 1 (1991).
- [25] R.A. Abramovitch, Organic Preparations and Procedures International., 23, 683 (1991).
- [26] a) A. Loupy, Spectra Analyses., 175, 33 and quoted references (1993); b) A. Loupy, G. Bram and J. Sansoulet, New J. Chem., 16, 233 (1992); c) G. Bram, A. Loupy, D. villemin, in "Solid supports and catalysts in organic synthesis", Edt Ellis Horwood, PTR. Frentice Hall, London, Chapter 12, 302 (1992).
- [27] R.A. Abramovitch, Organic, Prep. and Pmced Int., 23, 685 (1991) and references therein.
- [28] R. Gedye, F. Smith, K. Westaway, A. Humera, L. Baldisera and L. Laberge, Tetrahedron. Lett, 26, 279 (1986).
- [29] R.J. Giguere, T.L. Bray, S.M. Duncan and G. Majetich, *Tetrahedron. Lett.*, 27, 4945 (1986).
- [30] A.K. Bose, M.S. Manhas, M. Ghosh, V.S. Raju, K. Tabei and Urbanezyk., *Heterocycles.*, 30, 741 (1990).
- [31] (a) C.M. Duncan, *Tetrahedron Lett.*, 27 (41), 4945 (1986); b) A.K. Bose, M.S. Manhas, M. Ghosh et al., J. Org. Chem., 56, 6968 (1991).
- [32] B.K. Banik, M.S. Manhas, S.N. Newaz and A.K. Bose, Biorg. Med. Chem. Lett., 3, 2363 (1993).
- [33] K.C. Joshi, A. Dandia and S. Sanan, J. Fluorine, Chem., 44, 59 (1989).
- [34] K.C. Joshi, A. Dandia and S. Bhagat, J. Fluorine, Chem., 48, 169 (1990).
- [35] K.C. Joshi,, A. Dandia and N. Ahmed, Heterocycles., 24, 2479 (1986).
- [36] A. Dandia, V. Sehgal and P. Singh, Pharmazie., 49, 364 (1994).
- [37] A. Dandia, V. Sehgal and M. Upreti, Phosphorus, Sulfur and Silicon., 105, 93 (1995).
- [38] K.C. Joshi, A. Dandia and S. Baweja, Indian, J. Chem., 29 B, 766 (1990).
- [39] V. Sehgal, P. Singh, A. Dandia and R. Bohra, Acta Cryst., 1994.
- [40] A. Dandia, V Kaur and P. Singh, Indian, J. of Pharmaceutical Sci., 55 (4), 129(1993).
- [41] A. Dandia, and A. Kumari, Phosphorus, Sulfur and Silicon., 97, 27 (1994).