GOULD-JACOB TYPE OF REACTION IN THE SYNTHESIS OF THIENO[3,2-*e*]PYRIMIDO[1,2-*c*]PYRIMIDINES: A COMPARISON OF CLASSICAL HEATING *VS* SOLVENT-FREE MICROWAVE IRRADIATION

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Abstract- Gould-Jacob type of reaction for the synthesis of thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines (5) has been carried out conventionally by the condensation between 4-aminothieno[2,3-*d*]pyrimidines (2) and diethyl ethoxymethylenemalonate (3) *via* acyclic intermediates diethyl *N*-[5,6-disubstituted thieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonates (4) and the results obtained were compared with single step microwave technique under solvent free conditions for the synthesis of 5.

Diethyl ethoxymethylenemalonate (EMME) is widely used in the push-pull alkane,[1] 1,4-addition elimination,[2] 1,4-addition,[3] [3+2] cycloadditions,[4] protecting group for amino acids,[5] and Diels-Alder reactions.[6] EMME has also been employed frequently in Gould-Jacob reaction for the synthesis of quinoline derivatives.[7] Furthermore, a number of heterocyclic systems such as 1,8-naphtharidines, pyrazolinones, pyrones, xanthyrones, guanidine derivatives, 1,2,4-triazoles, 3-oxo-1,2,6-thiadiazines, 8-oxoimidazo[1,2-*a*]pyrimidines, 2*H*-pyrido[1,2-*a*]pyrimidin-4-ones, 3*H*-pyrrolo[1,2-*a*]indol-3-one derivatives and 1*H*-1,4-benzodiazepines have been prepared[8] using EMME as a synthon. Recently, Dave *et al.*[9] reported a novel route for the synthesis of pyrido[3,2-*e*]pyrimido[1,2-*c*]pyrimidines using the same synthon. Now-a-days, there is growing interest in the applications of microwave heating in organic chemistry[10-13] due to its remarkable advantages such as decrease in reaction time, solvent-free cleaner reactions, easier work-up, and better yields. The solvent-free reactions[14-16] under microwave conditions are especially appealing for providing an environmentally benign system. So far no attention has been given towards the synthesis of angu-

lar thienopyrimidopyrimidines using EMME as a synthon. Therefore, in continuation of our interest[17,18] in fused triheterocyclic systems we report herein the Gould-Jacob type of reaction for the synthesis of novel thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines (5) using conventional as well as microwave methodologies, and the comparative study of both the methods has been carried out.

Cyclocondensation of 4,5-disubstituted 2-amino-3-cyanothiophenes[19] (1) with formamide afforded the intermediates 5,6-disubstituted 4-aminothieno[2,3-*d*]pyrimidines[20] (2) required for the synthesis of thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines (5) (Scheme 1).

1 2

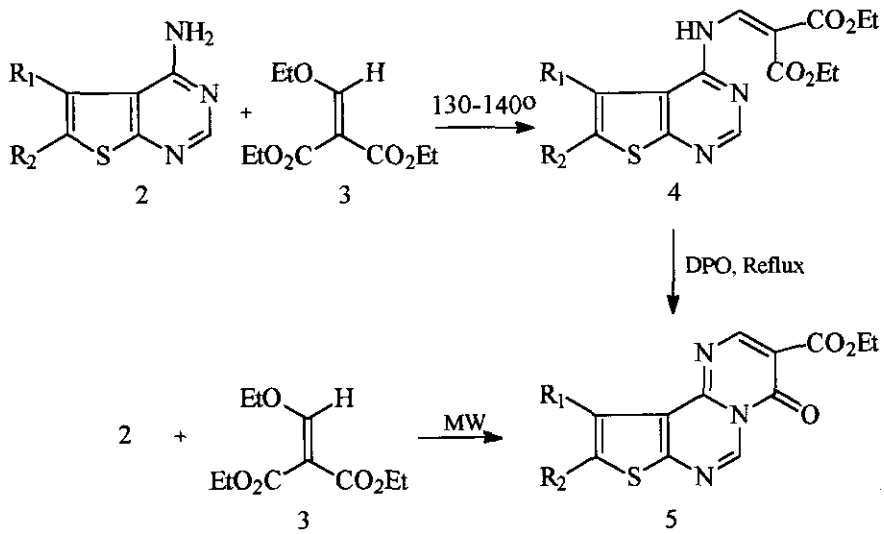
Scheme 1

In two step conventional method, 4-aminothieno[2,3-*d*]pyrimidines (2) were condensed with EMME (3) at 130-140 ^{0}C for 3.5-4.0 h to obtain diethyl *N*-[5,6-disubstituted thieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonates (4) which on thermal cyclization in boiling diphenyl oxide at 250 ^{0}C for 1.5-2.0 h provided 8,9-disubstituted thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines (5) in 42-61 % of overall yields from 4-aminothieno[2,3-*d*]pyrimidines (2) (Method A). On the other hand, single step microwave assisted reactions of 2 with EMME (3) without solvent (Method B) provided the identical compounds (5) within 7-10 min in 80-83 % yields (Scheme 2). Thus, the microwave assisted synthesis of thienopyrimidopyrimidines (5) has remarkable advantages over the conventional technique because of easier work-up, better yields, and rapid and solvent-free cleaner reactions. The comparison between conventional and microwave methodologies has been shown in the Table 1.

IR (KBr) spectra of 4 exhibited a characteristic band for NH in the region 3280-3260 cm^{-1} along with two sharp absorptions around 1700-1668 cm^{-1} due to carbonyl groups of two ester functionalities and the C=C and C=N vibrations were found at 1608-1500 cm^{-1}. The absence of amino vibrations in the region 3500-3400 cm^{-1} and the presence of absorption near 3256-3250 cm^{-1} due to NH suggested the formation of acyclic intermediates (4). ^{1}H NMR ($CDCl_3$) of 4 exhibited a doublet at δ 10.56-11.61 integrating for 1H because of the NH proton. A doublet for vinyl proton in the area δ 9.36-8.90 and a singlet at δ 8.60-8.77 due to pyrimidine ring proton were found to be present. Twin triplet and quartet in the region δ 1.31-1.51 and δ 4.33-4.51 integrating each for 3H and 2H respectively were responsible for two ethyl groups present in malonates (4). The absence of a band due to NH group in the area 3256-3350 cm^{-1} in the IR (KBr)

spectra supported the formation of angular thienopyrimidopyrimidines (5). Absorption at 1740-1724 cm^{-1} appeared due to an ester carbonyl group whereas an absorption due to lactone was found to be shifted 20-30 cm^{-1} to a higher wave number as compared to the ketones of acyclic malonates producing a sharp band in the region 1684-1680 cm^{-1}. The presence of triplet at δ 1.39-1.53 (3H) and quartet at δ 4.39-4.52 (2H) in the ^1H NMR ($CDCl_3$) spectra of 5 indicated the presence of a single ethyl group. Whereas pyrimidine protons at C2 and C6 were appeared as singlet at δ 6.97-7.60 and δ 8.85-9.05 each integrating for one proton.

In conclusion, we have developed a simple, fast, solvent-free and high yielding method for the synthesis of thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines.



4,5	R_1	R_2
a	-$(CH_2)_3$-	
b	-$(CH_2)_4$-	
c	Me	H
d	Me	Me
e	C_6H_5	Me
f	4-$OMeC_6H_4$	H

Scheme 2

Table 1. A comparison between conventional and microwave assisted synthesis of thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines (5)

Compound No.	Conventional (Method A)		Microwave[c] (Method B)		Melting point ^{0}C
	Time[a] (h)	Yield[b] %	Time (min)	Yield %	
5a	5.0	51	7.0	82	194-195
5b	5.5	61	7.5	83	221-222
5c	5.0	45	7.0	81	197-198
5d	6.0	54	8.0	80	172-173
5e	5,5	42	9.0	83	219-220
5f	6.0	44	10.0	82	168-169

[a]overall time on the basis of two steps, [b]overall yields on the basis of starting compounds (2).

[c]Microwave irradiation was carried out in a domestic microwave oven (BPL, BMO 700T).

EXPERIMENTAL

Melting points were determined by electrothermal method in an open capillary tube and are uncorrected. The IR spectra were recorded in cm^{-1} for KBr pellets on Buck scientific spectrophotometer. The ^{1}H NMR spectra were recorded on Varian 300 MHz spectrometer in dueteriochloroform using TMS as internal standard and the chemical shifts are expressed in δ ppm. MS spectra were recorded on LKB 9000 mass spectrometer. The purity of the compounds was routinely checked by TLC using silica gel G and the spots were exposed in iodine vapour.

The procedures given for the synthesis of 4 and 5 were utilized in the formation of compounds (4a-4f, and 5a-5f).

Method A: A two step conventional procedure for the synthesis of 8,9-disubstituted 3-carbethoxy-4-oxothieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines (5a-f) (General procedure)

Step 1: Diethyl *N*-[5,6-disubstituted thieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonates (4a-f) (General Procedure)

A mixture of 5,6-disubstituted 4-aminothieno[2,3-*d*]pyrimidines[20] (2) (0.01 mol) and EMME (3) (0.01 mol, 2.16 g) was heated at 130-140 ^{0}C for 3.5 to 4.0 h. The reaction mixture was allowed to attain the rt

and to this was added 25 mL of *n*-hexane. The solid thus obtained was filtered, dried and crystallized from ethanol.

Diethyl *N*-[cyclopenta[5,6]thieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonate (4a). yield : 79 %; mp : 172-173 ^{0}C; IR (KBr) : 3250 (NH); 3080-2930 (CH); 1690, 1670 (C=O); 1598-1504 (C=C, C=N) cm^{-1}; ^{1}H NMR ($CDCl_3$) : δ 1.51 (t x 2, J = 6.60 Hz, 6H, $CH_2C\underline{H}_3$), δ 2.73 (m, J = 6.80 Hz, 2H, $CH_2C\underline{H}_2CH_2$), δ 2.90 (t, J = 6.08 Hz, 2H, $C\underline{H}_2CH_2CH_2$), δ 3.20 (t, J = 5.70 Hz, 2H, $CH_2CH_2C\underline{H}_2$), δ 4.51 (q x 2, J = 7.71 Hz, 4H, $C\underline{H}_2CH_3$), δ 8.65 (s, 1H, Ar-H at C2), δ 9.22 (d, J = 12.35 Hz, 1H, vinyl-H), δ 11.60 (d, J = 11.80 Hz, 1H, NH); MS : 361 (M^+). Anal. Calcd for $C_{17}H_{19}N_3O_4S$: C, 56.49; H, 5.30; N, 11.62; S, 8.87. Found : C, 56.59; H, 5.48; N, 11.32; S, 9.01.

Diethyl *N*-[cyclohexa[5,6]thieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonate (4b). yield : 82 %; mp : 159-161 ^{0}C; IR (KBr) : 3255 (NH); 3085-2940 (CH); 1692, 1670 (C=O); 1594-1500 (C=C, C=N) cm^{-1}; ^{1}H NMR ($CDCl_3$) : δ 1.41 (t x 2, J = 6.90 Hz, 6H, $CH_2C\underline{H}_3$), δ 1.95 (m, J = 7.26 Hz, 4H, $CH_2C\underline{H}_2C\underline{H}_2CH_2$), δ 2.85 (t, J = 6.13 Hz, 2H, $C\underline{H}_2CH_2CH_2CH_2$), δ 3.14 (t, J = 5.68 Hz, 2H, $CH_2CH_2CH_2C\underline{H}_2$), δ 4.33 (q x 2, J = 7.20 Hz, 4H, $C\underline{H}_2CH_3$), δ 8.63 (s, 1H, Ar-H at C2), δ 9.36 (d, J = 12.03 Hz, 1H, vinyl-H), δ 11.61 (d, J = 11.88 Hz, 1H, NH); MS : 375 (M^+). Anal. Calcd for $C_{19}H_{21}N_3O_4S$: C, 57.58; H, 5.64; N, 11.19; S, 8.54. Found : C, 57.50; H, 5.40; N, 11.42; S, 8.77.

Diethyl *N*-[5-methylthieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonate (4c). yield : 70 %; mp : 170-171 ^{0}C; IR (KBr) : 3255 (NH); 3080-2950 (CH); 1696, 1668 (C=O); 1604-1504 (C=C, C=N) cm^{-1}; ^{1}H NMR ($CDCl_3$) : δ 1.39 (t x 2, J = 6.80 Hz, 6H, $CH_2C\underline{H}_3$), δ 2.40 (s, 3H, CH_3), δ 4.34 (q x 2, J = 7.10 Hz, 4H, $C\underline{H}_2CH_3$), δ 7.23 (s, 1H, Ar-H at C6), δ 8.60 (s, 1H, Ar-H at C2), δ 8.90 (d, J = 12.09 Hz, 1H, vinyl-H), δ 10.60 (d, J = 11.75 Hz, 1H, NH); MS : 335 (M^+). Anal. Calcd for $C_{15}H_{17}N_3O_4S$: C, 53.72; H, 5.11; N, 12.53; S, 9.56. Found : C, 53.49; H, 5.45; N, 12.32; S, 9.30.

Diethyl *N*-[5,6-dimethylthieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonate (4d). yield : 83 %; mp : 141-142 ^{0}C; IR (KBr) : 3250 (NH); 3090-2950 (CH); 1694, 1676 (C=O); 1600-1500 (C=C, C=N) cm^{-1}; ^{1}H NMR ($CDCl_3$) : δ 1.42 (t x 2, 6H, $CH_2C\underline{H}_3$, J = 6.80 Hz), δ 2.40 (s, 3H, CH_3), δ 2.45 (s, 3H, CH_3), δ 4.38 (q x 2, J = 7.18 Hz, 4H, $C\underline{H}_2CH_3$), δ 8.62 (s, 1H, Ar-H at C2), δ 8.95 (d, J = 12.15 Hz, 1H, vinyl-H), δ 10.61 (d, J = 11.80 Hz, 1H, NH); MS : 349 (M^+). Anal. Calcd for $C_{16}H_{19}N_3O_4S$: C, 55.00; H, 5.48; N, 12.03; S, 9.18. Found : C, 55.09; H, 5.40; N, 12.30; S, 8.89.

Diethyl *N*-[5-phenyl-6-methylthieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonate (4e). yield : 70 %; mp : 115-116 ^{0}C; IR (KBr) : 3254 (NH); 3050-2960 (CH); 1700, 1675 (C=O); 1600-1504 (C=C, C=N) cm^{-1}; ^{1}H NMR ($CDCl_3$) : δ 1.25-1.51 (t x 2, 6H, J = 6.98 Hz, $CH_2C\underline{H}_3$), δ 2.45 (s, 3H, CH_3), δ 4.52 (q x 2, J = 7.20 Hz, 4H, $C\underline{H}_2CH_3$), δ 7.00-7.35 (m, 5H, Ar-H), δ 8.65 (s, 1H, Ar-H at C2), δ 9.10 (d, J = 12.25 Hz,

1H, vinyl-H), δ 10.56 (d, J = 11.90 Hz, 1H, NH); MS : 411 (M^+). Anal. Calcd for $C_{21}H_{21}N_3O_4S$: C, 61.29; H, 5.14; N, 10.21; S, 7.79. Found : C, 61.49; H, 4.90; N, 9.99; S, 7.67.

Diethyl *N*-[5-(4-methoxyphenyl)thieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonate (4f). yield : 85 %; mp : 143-144 ^{0}C; IR (KBr) : 3256 (NH); 3060-2980 (CH); 1700, 1680 (C=O); 1608-1504 (C=C, C=N) cm^{-1}; ^{1}H NMR ($CDCl_3$) : δ 1.31 (t x 2, J = 6.57 Hz, 6H, CH_2CH_3), δ 3.88 (s, 3H, OCH_3), δ 4.21 (q x 2, J = 7.08 Hz, 4H, CH_2CH_3), δ 7.00-7.34 (m, 5H, Ar-H), δ 8.77 (s, 1H, Ar-H at C2), δ 9.27 (d, J = 12.30 Hz, 1H, vinyl-H), δ 10.57 (d, J =11.95 Hz, 1H, NH); MS : 427 (M^+). Anal. Calcd for $C_{21}H_{21}N_3O_5S$: C, 59.00; H, 4.95; N, 9.83; S, 7.50. Found : C, 59.04; H, 4.79; N, 10.01; S, 7.80.

Step 2 : 8,9-Disubstituted 3-carbethoxy-4-oxothieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines (5a-f) (General procedure)

Diethyl *N*-[5,6-disubstituted thieno[2,3-*d*]pyrimidin-4-yl]aminomethylenemalonates (4) (1.0 g) was dissolved in boiling diphenyl oxide (5 mL) and heated at 250 ^{0}C for 1.0-1.5 h. The excess of solvent was distilled *in vacuo* and methanol (20 mL) was added to the cooled reaction mixture, the solid thus obtained was filtered and crystallized from toluene.

Method B: Microwave assisted single step synthesis of 8,9-disubstituted 3-carbethoxy-4-oxothieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidines (5a-f) (General procedure)

A mixture of 5,6-disubstituted 4-aminothieno[2,3-*d*]pyrimidines (2) (0.01 mol) and EMME (3) (0.01 mol, 2.16 g) was taken in an open pyrex tube and subjected to microwave irradiation in a domestic microwave oven (BPL, BMO 700T) at an output of about 700 watts for the specified time as mentioned in Table 1. The reaction mixture was allowed to cool to rt and the solid obtained was crystallized from toluene. The products thus obtained were identical with the products formed by Method A which were confirmed on the basis of TLC, mp, mmp, elemental and spectral analysis. The yields and melting points are given in Table 1.

3-Carbethoxy-4-oxocyclopenta[8,9]thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidine (5a). IR (KBr) :3090-2992 (CH); 1736, 1680 (C=O); 1588-1492 (C=C, C=N) cm^{-1}; ^{1}H NMR ($CDCl_3$) : δ 1.51 (t, J=6.72 Hz, 3H, CH_2CH_3), δ 2.80 (m, J = 6.48 Hz, 2H, $CH_2CH_2CH_2$), δ 2.95 (t, J = 5.75 Hz, 2H, $CH_2CH_2CH_2$), δ 3.35 (t, 2H, J = 5.62 Hz, $CH_2CH_2CH_2$), δ 4.51 (q, J = 6.96 Hz, 2H, CH_2CH_3), δ 7.24 (s, 1H, Ar-H at C2), δ 8.95 (s, 1H, Ar-H at C6); MS : 315 (M^+). Anal. Calcd for $C_{15}H_{13}N_3O_3S$: C, 57.13; H, 4.16; N, 13.38; S, 10.17. Found : C, 57.09; H, 4.30; N, 13.09; S, 10.34.

3-Carbethoxy-4-oxocyclohexa[8,9]thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidine (5b). IR (KBr) : 3090-2992 (CH); 1740, 1680 (C=O); 1588-1492 (C=C, C=N) cm^{-1}; ^{1}H NMR ($CDCl_3$) : δ 1.53 (t, J = 6.62 Hz, 3H,

CH_2CH_3), δ 1.94 (m, J = 6.55 Hz, 4H, $CH_2CH_2CH_2CH_2$), δ 2.95 (t, J = 4.10 Hz, 2H, $CH_2CH_2CH_2CH_2$), δ 3.26 (t, J = 4.60 Hz, 2H, $CH_2CH_2CH_2CH_2$), δ 4.42 (q, J = 7.21 Hz, 2H, CH_2CH_3), δ 7.27 (s, 1H, Ar-H at C2), δ 9.05 (s, 1H, Ar-H at C6); MS : 329 (M^+). Anal. Calcd for $C_{16}H_{15}N_3O_3S$: C, 58.34; H, 4.59; N, 12.76; S, 9.74. Found : C, 58.07; H, 4.30; N, 12.90; S, 9.39.

3-Carbethoxy-4-oxo-9-methylthieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidine (5c). IR (KBr) : 3080-2990 (CH); 1728, 1684 (C=O); 1592-1492 (C=C, C=N) cm^{-1}; ^1H NMR ($CDCl_3$) : δ 1.40 (t, 3H, J = 6.73 Hz, CH_2CH_3), δ 2.40 (s, 3H, CH_3), δ 4.41 (q, J = 7.10 Hz, 2H, CH_2CH_3), δ 7.20 (s, 1H, Ar-H at C2); δ 7.35 (s, 1H, Ar-H at C8), δ 8.90 (s, 1H, Ar-H at C6); MS : 289 (M^+). Anal. Calcd for $C_{13}H_{11}N_3O_3S$: C, 53.97; H, 3.83; N, 14.52; S, 11.08. Found : C, 54.08; H, 3.60; N, 14.80; S, 11.36.

3-Carbethoxy-4-oxo-8,9-dimethylthieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidine (5d). IR (KBr) : 3080-2990 (CH); 1728, 1680 (C=O); 1592-1490 (C=C, C=N) cm^{-1}; ^1H NMR ($CDCl_3$) : δ 1.25-1.44 (t, J = 6.70 Hz, 3H, CH_2CH_3), δ 2.30 (s, 3H, CH_3), 2.50 (s, 3H, CH_3), δ 4.44 (q, J = 7.20 Hz, 2H, CH_2CH_3), δ 7.21 (s, 1H, Ar-H at C2), δ 8.91 (s, 1H, Ar-H at C6); MS : 303 (M^+). Anal. Calcd for $C_{14}H_{13}N_3O_3S$: C, 55.43; H, 4.32; N, 13.85; S, 10.57. Found : C, 55.18; H, 4.40; N, 13.80; S, 10.37.

3-Carbethoxy-4-oxo-8-methyl-9-phenylthieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidine (5e). IR (KBr) : 3070-2980 (CH); 1724, 1680 (C=O); 1592-1494 (C=C, C=N) cm^{-1}; ^1H NMR ($CDCl_3$) : δ 1.52 (t, J = 6.80 Hz, 3H, CH_2CH_3), δ 2.45 (s, 3H, CH_3), δ 4.50 (q, J = 7.20 Hz, 2H, CH_2CH_3), δ 7.06-7.46 (m, 6H, Ar-H), δ 8.85 (m, 1H, Ar-H at C6); MS : 365 (M^+). Anal. Calcd for $C_{19}H_{15}N_3O_3S$: C, 62.45; H, 4.14; N, 11.50; S, 8.78. Found : C, 62.26; H, 3.88; N, 11.85; S, 8.43.

3-Carbethoxy-4-oxo-9-(4-methoxyphenyl)thieno[3,2-*e*]pyrimido[1,2-*c*]pyrimidine (5f). IR (KBr) : 3070-2980 (CH); 1728, 1684 (C=O); 1590-1492 (C=C, C=N) cm^{-1}; ^1H NMR ($CDCl_3$) : δ 1.39 (t, J = 6.88 Hz, 3H, CH_2CH_3), δ 3.89 (s, 3H, OCH_3), δ 4.41 (q, J = 7.29 Hz, 2H, CH_2CH_3), δ 6.97-7.49 (m, 6H, Ar-H), δ 8.87 (m, 1H, Ar-H at C6); MS : 381 (M^+). Anal. Calcd for $C_{19}H_{15}N_3O_4S$: C, 59.83; H, 3.96; N, 11.02; S, 8.41. Found : C, 59.28; H, 4.08; N, 11.36; S, 8.33.

ACKNOWLEDGMENT

We wish to thank Regional Sophisticated Instrumental Center, Central Drug Research Institute, Lucknow, India for H^1 NMR and mass spectral analysis.

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Received, 15th February, 1999