7. ABSTRACT OF INVENTION:

This invention describes the synthesis of 1, 3-diaryl-(1H, 3H)-6-methylpyrimidine-2, 4-diones by *Chapman rearrangement* of 2, 4-diaryloxy 6-methylpyrimidines under microwave irradiation.

5. CLAIMS:

We Claim:

1. Synthesis of 1, 3-di-(2-methylphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4a)** as described in **Example-1** by *Chapman rearrangement* of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** under microwave irradiation (900W) in 16 minutes.

2. Synthesis of 1, 3-di-(2-carbethoxyphenyl)-(1H, 3H)-6-methylpyrimidine -2, 4-dione **(4b)** as described in **Example-2** by *Chapman rearrangement* of 2, 4-di-(2-carbethoxyphenoxy)-6-methylpyrimidine **(3b)** under microwave irradiation (900W) in 18 minutes.

3. Synthesis of 1, 3-di-(4-carbmethoxyphenyl)-(1H, 3H)-6-methylpyrimidine **-2, 4-dione (4c)** as described in **Example-3** by *Chapman rearrangement* of 2, 4-di-(4-carbmethoxyphenoxy)-6-methylpyrimidine **(3c)** under microwave irradiation (900W) in 13 minutes.

4. Synthesis of 1, 3-di-(2-naphthyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4d)** as described in **Example-4** by *Chapman rearrangement* of 2, 4-di-(2-naphthoxy)-6-methylpyrimidine **(3d)** under microwave irradiation (900W) in 17 minutes.

5. Synthesis of 1, 3-di-(4-carbethoxy-2, 6-dimethoxyphenyl)-(1H, 3H)-6-methyl pyrimidine-2, 4-dione **(4e)** as described in **Example-5** by *Chapman rearrangement of* 2, 4-di-(4-carbethoxy-2, 6-dimethoxyphenoxy)-6-methylpyrimidine **(3e)** under microwave irradiation (900W) in 18 minutes.

6. Synthesis of 1, 3-di-(2-ethoxyphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4f)** as described in **Example-6** by *Chapman rearrangement* of 2, 4-di-(2-ethoxyphenoxy)-6-methylpyrimidine **(3f)** under microwave irradiation (900W) in 20 minutes.

7. Synthesis of 1, 3-di-(4-chloro-3, 5-dimethylphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4g)** as described in **Example-7** by *Chapman rearrangement* of 2, 4-di-(4-chloro-3, 5-dimethylphenoxy)-6-methylpyrimidine **(3g)** under microwave irradiation (900W) in 15 minutes.

8. Synthesis of 1, 3-diphenyl-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4h)** as described in **Example-8** by *Chapman rearrangement* of 2, 4-diphenoxy-6-methylpyrimidine **(3h)** under microwave irradiation (900W) in 16 minutes.

9. 2, 4-diaryloxy-6-methylpyrimidines **(3a-3h)** underwent facile *Chapman rearrangement* under microwave irradiation to afford 1, 3- disubstituted-6-methylpyrimidine-2, 4-diones **(4a-4j)** which provides a simpler and environmental friendly procedure. Thus, microwave is a convenient way towards the goal of green, sustainable chemistry, and is strongly recommended to use in organic preparations.

6. DATE: 29th **October 2013**

SIGNATURES:

Dr. M. M. V. Ramana **Dr. Sanjay C. Pawar**

4. DESCRIPTION:

Title:

A process for the preparation of 1, 3-diaryl-(1H, 3H)-6-methylpyrimidine-2, 4-diones under microwave irradiation.

Field of invention:

This invention describes the synthesis of 1, 3-diaryl-(1H, 3H)-6-methylpyrimidine-2, 4-diones by *Chapman rearrangement* of 2, 4-diaryloxy-6-methylpyrimidines under microwave irradiation.

Background of invention and Prior art:

1, 3- disubstituted pyrimidine-2, 4-diones possess biological activity and have good pharmaceutical applications. These are well known in prior arts including EP 1986651 A1, WO 1998018781 A3, US 6232318 B1, US 6420385. The most prominent representatives are 5-fluoro-(1H, 3H)-pyrimidine-2, 4-dione and 5-Methyl-(1H, 3H)-pyrimidine-2, 4-dione derivatives which are widely used in therapy, mainly as antiviral and antineoplastic agents. (Ch. Hoffmann, J. K. Rockstroh and B.S. Kamps, *HIV Medicine*, **2005**, Flying Publisher, Steinhauser, Paris, pp. 620-683) 5-[bis(2-chloroethyl)amino]-1H-pyrimidine-2, 4-dione, 5-[N, N'-bis-(2'-chloroethyl)-amino] pyrimidine-2, 4-dione, is used orally in the treatment of several leukemias, and 5-nitro-(1H, 3H)-pyrimidine-2, 4-dione derivatives exhibit macrophage growth inhibition. (P. G. Baraldi, R. Romagnoli, A. E. Guadix, Pinada de las Infantas, M. A. Gallo, A. Espinosa, A. Martinez, J. P. Bingham and J. A. Hartey, *J. Med. Chem.*, **2002**, 45, 3630-3638) Some substituted pyrimidine-2, 4-dione are also useful intermediate in the synthesis of other 1H, 3H-pyrimidine-2, 4-dione derivatives. (A. Gondela and K. Walczak, *Tetrahedron Lett.* **2003**, 44,7291-7293) Some pyrimidinedione derivatives are used for treating dyslipidemia, metabolic disorders, cancer, diabetic complications, inflammation, respiratory diseases, gastroenterological diseases, hematological diseases etc. (WO 2008127591 A2) Some pyrimidinedione derivatives are well known for their use as pesticides, herbicides, and insecticides. Such materials are well known in the prior arts including WO 1998008824 A1, US 5344812, US 5674810, WO 9952906, JP 03287585 A2.

6-methyl-1, 3-oxazine-2, 4-(3H)-dione when treated with an excess of arylamines was transformed into the appropriate 1-aryl-6-methyl pyrimidine-2, 4-diones. (H. Singh, P. Singh, P. Aggarwal and S. Kumar, *J. Chem. Soc., Perkin Trans.* 1, **1993**, 731-735). Pyrimidine-2, 4-dione derivatives, in reactions with diarylidonium salts, gave the appropriate N-mono- and N,

N'-diarylation products with high regioselectivity. (T. Maruyama, K. Fujiwara and M. Fukuhara, *J. Chem. Soc., Perkin Trans.* 1, **1995**, 733-734).

Although most of the above methodologies have their own synthetic values, some limitations mainly due to the long reaction times, harsh reaction conditions, tedious preparation procedures, use of solvents etc.; which could represent significant drawbacks for preparative purposes. Thus, there is scope for the synthesis of them by simple and eco-friendly method. The present invention describes the application of microwave irradiation for the synthesis of 1, 3-diaryl-(1H, 3H)-6-methylpyrimidine-2, 4-diones in two steps through *Chapman rearrangement* under microwave irradiation in absence of solvent in second step.

Microwave irradiation is one of the most promising non conventional methodologies used in organic synthesis. Use of microwave generally allows to conduct organic reactions in an easy way which also dramatically decreases reaction time, clean work up and better reaction yield with high purity. (D. M. P. Mingos, D. R. Baghurst, *Chem. Soc. Rev.* 20 , **1991**, 1-47)

Description of the invention:

The main objective of the present invention is to synthesize compounds with (1) minimum steps of reactions, (2) minimum use of solvents, (3) lesser reaction time, (4) clean work up, (5) better efficiency of product.

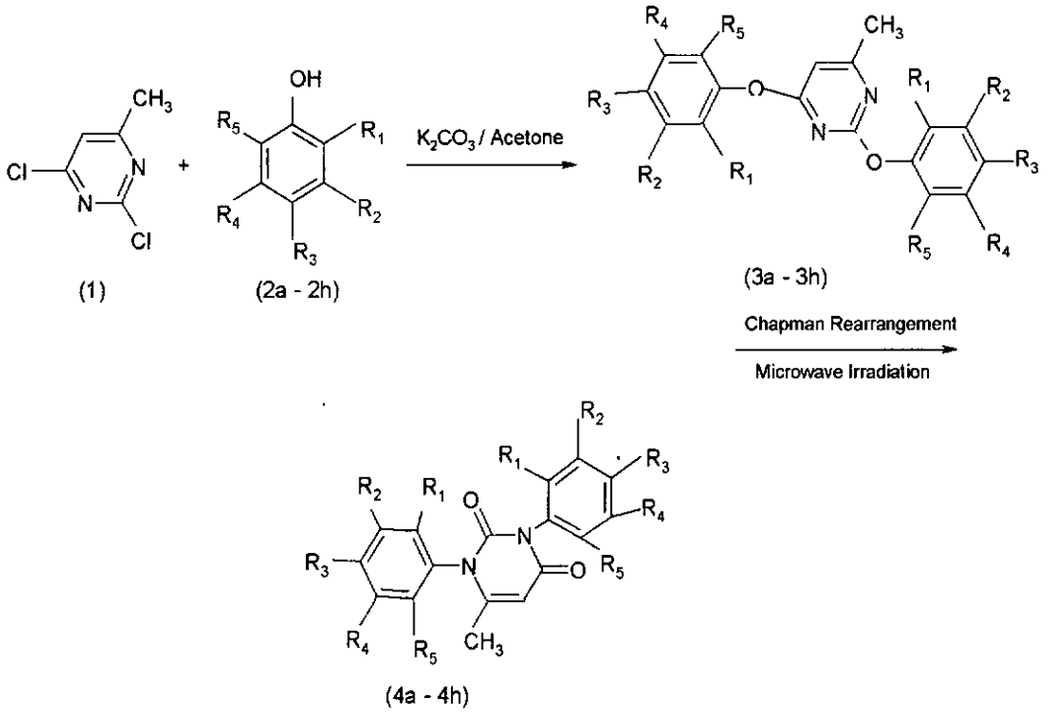
The present invention describes synthesis of 1, 3-diaryl-(1H, 3H)-6-methylpyrimidine-2, 4-diones in two steps via *Chapman rearrangement* of corresponding 2, 4-diaryloxy-6-methylpyrimidines under microwave irradiation under solvent free condition.

The present invention is successful in attaining all the above objectives providing a simpler and environmental friendly alternative for the conventional procedures.

The thermal rearrangement of aryl imidates to N, N-diaryl amides is generally known as the *Chapman rearrangement.* (Chapman, *J. Chem. Soc.* **1925**, 569; Chapman, *J. Chem. Soc.* **1927**, 1743)

2, 4-dichloro-6-methylpyrimidine **(1)** was condensed with various phenols. This yielded the respective 2, 4-diaryloxy-6-methylpyrimidines **(3a-3j)**. These were then subjected to *Chapman rearrangement* under microwave irradiation in absence of solvent to afford the corresponding 1, 3-diaryl-(1H, 3H)-6-methylpyrimidine-2, 4-diones. **(Scheme)**

Scheme:



Compounds	R_1	R_2	R_3	R_4	R_5
2a, 3a, 4a	CH_3	H	H	H	H
2b, 3b, 4b	$COOC_2H_5$	H	H	H	H
2c, 3c, 4c	H	H	$COOCH_3$	H	H
2d, 3d, 4d	H	H			H
2e, 3e, 4e	OCH_3	H	$COOC_2H_5$	H	OCH_3
2f, 3f, 4f	OC_2H_5	H	H	H	H
2g, 3g, 4g	H	CH_3	Cl	CH_3	H
2h, 3h, 4h	H	H	H	H	H

Examples:

Example-1: Preparation of 1, 3-di-(2-methylphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione (4a).

Step A- Preparation of 2, 4-di-(2-methylphenoxy) -6-methylpyrimidine (3a).

A solution of 2, 4-dichloro-6-methylpyrimidine **(1)** (0.04M) in dry acetone (25ml) was added dropwise to a mixture of 2-methylphenol **(2a)** (0.08M) and K_2CO_3 (0.08M) in dry acetone (50 ml). After complete addition, the reaction mixture was slowly refluxed for 4 hours under dry conditions. Acetone was recovered by flash distillation, after completion of the reaction (TLC). The reaction mass was cooled to room temperature and poured in water (75ml) under stirring. It was extracted in dichloromethane (2 x 50ml) followed by washing with 5% aqueous solution of NaOH (2 x 25ml). The combined extracts were given water washing (2 x 25ml) and dried over sodium sulphate. Recovery of solvent afforded a solid which was filtered and recrystallised from ligroin to give off-white crystals (Yield: 58%) of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)**. m.p.: 106°C

Molecular formula: $C_{19}H_{18}N_2O_2$. Elemental analysis: Calculated: C (74.51%), H (5.88%), N (9.15%). Found: C (74.58%), H (5.75%), N (9.21%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 2.2(s, 9H), 6.3-7.0(m, 9H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 19-24, 124-169.

IR (KBr, cm^{-1}): 1240, 1345, 1609, 2976-3040.

HRMS: *m/z* cal. mass for $C_{19}H_{18}N_2O_2$ $[M+H]^+$=306.3602, obs. mass $[M+H]^+$= 306.3653.

Step B- Preparation of compound 4a.

In a flask, equipped with a condenser 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** (0.03M) was irradiated in microwave reactor (900W) for 16 minutes. The reaction mass was cooled to room temperature and treated with ligroin (50 ml). The solution was filtered. The filtrate on recovery of solvent afforded an oil of compound **4a**. (Yield: 71%)

Molecular formula: $C_{19}H_{18}N_2O_2$. Elemental analysis: Calculated: C (74.51%), H (5.88%), N (9.15%). Found: C (74.42%), H (5.92%), N (8.99%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.5(s, 6H), 1.8(s, 3H), 6.1- 7.5(m, 9H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 16-18, 100, 142, 126.5-136, 152, 164.

IR (KBr, cm^{-1}): 1343, 1607, 1641, 1683, 1694, 2943-3017.

HRMS: *m/z* cal. mass for $C_{19}H_{18}N_2O_2$ $[M+H]^+$=306.3602, obs. mass $[M+H]^+$= 306.3679.

Example-2: Preparation of 1, 3-di-(2-carbethoxyphenyl)-(1H, 3H)-6-methylpyrimidine-

2, 4-dione (4b).

Step A- Preparation of 2, 4-di-(2-carbethoxyphenoxy)-6-methylpyrimidine (3b).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloro-6-methylpyrimidine **(1)** and 2-carbethoxyphenol **(2b)** instead of 2-methylphenol **(2a)**, whereby **3b** was obtained as white crystals (Yield: 63%) after purification. (mp.:104°C)

Molecular formula: $C_{23}H_{22}N_2O_6$. Elemental analysis: Calculated: C (65.40%), H (5.21%), N (6.64%). Found: C (65.49%), H (5.16%), N (6.55%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.5-1.4(t, J=7.5 Hz, 6H), 2.4(s, 3H), 4.1-4.4(q, J=7.8 Hz, 4H), 6.4-7.8(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 11-17, 59-64, 119-169, 167.5-171.

IR (KBr, cm^{-1}): 1237, 1350, 1428, 1605, 1720, 1750, 2854-2095.

HRMS: *m/z* cal. mass for $C_{23}H_{22}N_2O_6$ $[M+H]^+$ =422.4328, obs. mass $[M+H]^+$ =422.4300.

Step B- Preparation of compound 4b.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(2-carbethoxyphenoxy)-6-methylpyrimidine **(3b)** instead of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** was irradiated in microwave reactor for 18 minutes, whereby **4b** was obtained as white crystals (Yield: 66%) after purification. (m.p.:144°C)

Molecular formula: $C_{23}H_{22}N_2O_6$. Elemental analysis: Calculated: C (65.40%), H (5.21%), N (6.64%). Found: C (65.47%), H (5.27%), N (6.71%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.2-1.4(t, J=7.1 Hz, 6H), 2.2(s, 3H), 4.05-4.4(q, J=7.7 Hz, 4H), 6.4-7.7(m, 9H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 12-14, 61-63.5, 108, 127-161, 140, 154, 167, 169-175.5.

IR (KBr, cm^{-1}): 1210, 1360, 1605, 1653, 1678, 1710, 1732, 2998-3000.

HRMS: *m/z* cal. mass for $C_{23}H_{22}N_2O_6$ $[M+H]^+$ =422.4328, obs. mass $[M+H]^+$ =422.4361.

Example-3: Preparation of 1, 3-di-(4-carbmethoxyphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione (4c).

Step A- Preparation of 2, 4-di-(4-carbmethoxyphenoxy)-6-methylpyrimidine (3c).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloro-6-methylpyrimidine **(1)** and 4-carbmethoxyphenol **(2c)** instead of 2-methylphenol **(2a)**, whereby **3c** was obtained as viscous oil (Yield: 56%) after purification.

Molecular formula: $C_{21}H_{18}N_2O_6$. Elemental analysis: Calculated: C (63.96%), H (4.57%), N (7.11%). Found: C (63.88%), H (4.65%), N (7.17%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 2.2(s, 3H), 3.8(s, 6H), 6.3-7.6(m, 9H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 22, 51-53, 125.5-171.5, 167-172.

IR (KBr, cm^{-1}): 1215, 1243, 1610, 1730, 2870-2957.

HRMS: *m/z* cal. mass for $C_{21}H_{18}N_2O_6$ $[M+H]^+$ =394.3794, obs. mass $[M+H]^+$ = 394.3719.

Step B- Preparation of compound 4c.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(4-carbmethoxyphenox-6-methylpyrimidine **(3c)** instead of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** was irradiated in microwave reactor for 13 minutes, whereby **4c** was obtained as viscous oil (Yield: 71%) after purification.

Molecular formula: $C_{21}H_{18}N_2O_6$. Elemental analysis: Calculated: C (63.96%), H (4.57%), N (7.11%). Found: C (64.05%), H (4.62%), N (7.15%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 2.2(s, 3H), 3.9(s, 6H), 6.3-7.2(m, 9H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 51-52.5, 100, 129-158, 140, 153.5, 165, 163.5-166.

IR (KBr, cm^{-1}): 1221, 1347, 1608, 1634, 1680, 1685, 1700, 2955.

HRMS: *m/z* cal. mass for $C_{21}H_{18}N_2O_6$ $[M+H]^+$ =394.3794, obs. mass $[M+H]^+$ = 394.3751.

Example-4: Preparation of 1, 3-di-(2-naphthyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione (4d).

Step A- Preparation of 2, 4-di-(2-naphthoxy)-6-methylpyrimidine (3d).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloro-6-methylpyrimidine **(1)** and 2-naphthol **(2d)** instead of 2-methylphenol **(2a)**, whereby **3d** was obtained as white crystals (Yield: 55%) after purification. (m.p.:113°C)

Molecular formula: $C_{25}H_{18}N_2O_2$. Elemental analysis: C (79.37%), H (4.76%), N (7.41%). Found: C (79.28%), H (4.84%), N (7.33 %).

^{1}H NMR (300 MHz, $CDCl_3$): δ 2.3(s, 3H), 6.2-7.20(m, 15H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 21, 123-172.

IR (KBr, cm^{-1}): 124, 1600, 2955-3005.

HRMS: *m/z* cal. mass for $C_{25}H_{18}N_2O_2$ $[M+H]^+$ =378.4248, obs. mass $[M+H]^+$=378.4201.

Step B- Preparation of compound 4d.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(2-naphthoxy)-6-methylpyrimidine **(3d)** instead of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** was irradiated in microwave reactor for 17 minutes, whereby **4d** was obtained as an oil (Yield: 69%) after purification.

Molecular formula: $C_{25}H_{18}N_2O_2$. Elemental analysis: C (79.37%), H (4.76%), N (7.41%). Found: C (79.45%), H (4.65%), N (7.51%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 2.0(s, 3H), 6-7.4(m, 15H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 21, 123-133, 111-142, 153, 164.

IR (KBr, cm^{-1}): 1335, 1608, 1640, 1682, 1689, 2874-2989.

HRMS: *m/z* cal. mass for $C_{25}H_{18}N_2O_2$ $[M+H]^+$ =378.4248, obs. mass $[M+H]^+$=378.4211.

Example-5: Preparation of 1, 3-di-(4-carbethoxy-2, 6-dimethoxyphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione (4e).

Step A- Preparation of 2, 4-di-(4-carbethoxy-2, 6-dimethoxyphenoxy)-6-methylpyrimidine (3e).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloro-6-methylpyrimidine **(1)** and ethyl-4-hydroxy-3, 5-dimethoxybenzoate (ethyl syringate) **(2e)** instead of 2-methylphenol **(2a)**, whereby **3e** was obtained as cream coloured crystals (Yield: 53%) after purification. (m.p.:112°C)

Molecular formula: $C_{27}H_{30}N_2O_{10}$ Elemental analysis: Calculated: C (59.78%), H (5.54%), N (5.17%). Found: C (59.89%), H (5.61%), N (5.21%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.4(t, J=6.9 Hz, 6H), 2.1(s, 3H), 3.9(s, 12H), 4.5(q, J= 7.2 Hz, 4H), 6.4-7.5(m, 5H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 19-23, 54, 56.5, 166-170, 127-174.

IR (KBr, cm^{-1}): 1198, 1238, 1339, 1612, 1710, 2854-2985.

HRMS: *m/z* cal. mass for $C_{27}H_{30}N_2O_{10}$ $[M+H]^+$ =542.5372, obs. mass $[M+H]^+$=542.5316

Step B- Preparation of compound 4e.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(4-carbethoxy-2, 6-dimethoxyphenoxy)-6-methylpyrimidine (3e) instead of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** was irradiated in microwave reactor for 18 minutes, whereby **4e** was obtained as an oil (Yield: 65%) after purification.

Molecular formula: $C_{27}H_{30}N_2O_{10}$ Elemental analysis: Calculated: C (59.78%), H (5.54%), N (5.17%). Found: C (59.87%), H (5.47%), N (5.29%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.4(t, J = 7.1Hz, 6H), 2.3(s, 3H), 3.8(s, 12H), 4.38(q, J = 7.5 Hz, 4H), 6.2-7.5 (m, 5H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 19.5-22, 53.5-55.4, 59-62, 103, 127.5-156, 140, 154, 162, 163-165.

IR (KBr, cm^{-1}): 1200, 1244, 1341, 1600, 1640, 1680, 1691, 1715, 2854-2998.

HRMS: *m/z* cal. mass for $C_{27}H_{30}N_2O_{10}$ $[M+H]^+$ =542.5372, obs. mass $[M+H]^+$=542.5305

Example-6: Preparation of 1, 3-di-(2-ethoxyphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione (4f).

Step A- Preparation of 2, 4-di-(2-ethoxyphenoxy)-6-methylpyrimidine (3f).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloro-6-methylpyrimidine **(1)** and 2-ethoxyphenol **(2f)** instead of 2-methylphenol **(2a)**, whereby **3f** was obtained as yellow crystals (Yield: 59%) after purification. (m.p.: 92°C)

Molecular formula: $C_{21}H_{22}N_2O_4$. Elemental analysis: Calculated: C (68.85%), H (6.01%); N (7.65%). Found: C (68.91%), H (5.89%), N (7.69%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.3(t, J=6.9 Hz, 6H), 2.4 (s, 3H), 4.1 (q, J=7.8Hz, 4H), 6.1-7.5 (m, 9H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 14.5-16, 58-63, 123-167.

IR (KBr, cm^{-1}): 1232, 1345, 1608, 2834-2975.

HRMS: *m/z* cal. mass for $C_{21}H_{22}N_2O_4$ $[M+H]^+$=366.4124, obs. mass $[M+H]^+$ = 366.4180.

Step B- Preparation of compound 4f.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(2-ethoxyphenoxy)-6-methylpyrimidine **(3f)** instead of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** was irradiated in microwave reactor for 20 minutes, whereby **4f** was obtained as an oil (Yield: 71%) after purification.

Molecular formula: $C_{21}H_{22}N_2O_4$. Elemental analysis: Calculated: C (68.85%), H (6.01%), N (7.65%). Found: C (68.96%), H (6.12%), N (7.71%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.5(t, J=7.8Hz, 6H), 2.0(s, 3H), 4.1(q, J=7.8Hz, 4H), 6.2-7.3(m, 9H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 13-15, 54.5-58.5, 102, 127-158, 142, 154, 168.

IR (KBr, cm^{-1}): 1240, 1349, 1614, 1640, 1680, 1691, 2854-2955.

HRMS: *m/z* cal. mass for $C_{21}H_{22}N_2O_4$ $[M+H]^+$=366.4124, obs. mass $[M+H]^+$ = 366.4155.

Example-7: Preparation of 1, 3-di-(4-chloro-3, 5-dimethylphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione (4g).

Step A- Preparation of 2, 4-di-(4-chloro-3, 5-dimethylphenoxy)-6-methylpyrimidine(3g).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloro-6-methylpyrimidine **(1)** and 4-chloro-3, 5-dimethylphenol **(2g)** instead of 2-methylphenol **(2a)**, whereby **3g** was obtained as colourless crystals (Yield: 62%) after purification. (m.p.: 114°C)

Molecular formula: $C_{21}H_{20}N_2O_2Cl_2$. Elemental analysis: Calculated: C (62.53%), H 4.96%), N (6.95%), Cl (17.62%). Found: C (62.59%), H (5.02%), N (7.01%), Cl (17.69%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 2.3(s, 3H), 2.5(s, 12H), 6.2-7.2(m, 5H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 16.5-23, 123-171.

IR (KBr, cm^{-1}): 774, 1233, 1352, 1610, 2980-3012.

HRMS: *m/z* cal. mass for $C_{21}H_{20}N_2O_2Cl_2$ $[M+H]^+$=403.3040, obs. mass $[M+H]^+$= 403.3074.

Step B- Preparation of compound 4g.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(4-chloro-3, 5-dimethylphenoxy)-6-methylpyrimidine **(3g)** instead of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** was irradiated in microwave reactor for 15 minutes, whereby **4g** was obtained as a solid (Yield: 69%) after purification. (m.p.: 153°C)

Molecular formula: $C_{21}H_{20}N_2O_2Cl_2$. Elemental analysis: Calculated: C (62.53%), H 4.96%), N (6.95%), Cl (17.62%). Found: C (62.45%), H (5.05%), N (6.85%), Cl (17.53%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.4(s, 12H), 1.8(s, 3H), 6.2-7.5(m, 5H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 16- 20, 101, 125-136, 144, 154, 167.

IR (KBr, cm^{-1}): 784, 1356, 1611, 1632, 1684, 1695, 2876-2955.

HRMS: *m/z* cal. mass for $C_{21}H_{20}N_2O_2Cl_2$ $[M+H]^+$ =403.3040, obs. mass $[M+H]^+$ = 403.3091.

Example-8: Preparation of 1, 3-diphenyl-(1H, 3H)-6-methylpyrimidine-2, 4-dione (4h).

Step A- Preparation of 2, 4-diphenoxy-6-methylpyrimidine (3h).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloro-6-methylpyrimidine **(1)** and phenol **(2h)** instead of 2-methylphenol **(2a)**, whereby **3h** was obtained as white crystals (Yield: 69%) after purification. (m.p.: 97°C) (Ghosh, S.K.; Dolly, R. S.; Mukherjee, M.K. *J. Med. Chem.* **1968**, *11 (6)*, 1237-8)

Step B- Preparation of compound 4h.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-diphenoxy-6-methylpyrimidine **(3h)** instead of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** was irradiated in microwave reactor for 16 minutes, whereby **4h** was obtained as white crystals (Yield: 65%) after purification. (m.p.:109°C)

Molecular formula: $C_{17}H_{14}N_2O_2$. Elemental analysis: Calculated: C (73.38%), H (5.04%), N (10.07%). Found: C (73.47%), H (4.96%), N (10.11%).

^{1}H NMR (300 MHz, $CDCl_3$): δ 2.4(s, 3H), 6.1-7.7(m, 11H)

^{13}C NMR (75 MHz, $CDCl_3$): δ 121-136, 110-140, 151, 162.

IR (KBr, cm^{-1}): 1335, 1608, 1640, 1682, 1689, 2874-2989.

HRMS: *m/z* cal. mass for $C_{17}H_{14}N_2O_2$ $[M+H]^+$ = 278.3068, obs. mass $[M+H]^+$ = 278.3007.

5. CLAIMS:

We Claim:

1. Synthesis of 1, 3-di-(2-methylphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4a)** as described in **Example-1** by *Chapman rearrangement* of 2, 4-di-(2-methylphenoxy)-6-methylpyrimidine **(3a)** under microwave irradiation (900W) in 16 minutes.

2. Synthesis of 1, 3-di-(2-carbethoxyphenyl)-(1H, 3H)-6-methylpyrimidine -2, 4-dione **(4b)** as described in **Example-2** by *Chapman rearrangement* of 2, 4-di-(2-carbethoxyphenoxy)-6-methylpyrimidine **(3b)** under microwave irradiation (900W) in 18 minutes.

3. Synthesis of 1, 3-di-(4-carbmethoxyphenyl)-(1H, 3H)-6-methylpyrimidine **-2, 4-dione (4c)** as described in **Example-3** by *Chapman rearrangement* of 2, 4-di-(4-carbmethoxyphenoxy)-6-methylpyrimidine **(3c)** under microwave irradiation (900W) in 13 minutes.

4. Synthesis of 1, 3-di-(2-naphthyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4d)** as described in **Example-4** by *Chapman rearrangement* of 2, 4-di-(2-naphthoxy)-6-methylpyrimidine **(3d)** under microwave irradiation (900W) in 17 minutes.

5. Synthesis of 1, 3-di-(4-carbethoxy-2, 6-dimethoxyphenyl)-(1H, 3H)-6-methyl pyrimidine-2, 4-dione **(4e)** as described in **Example-5** by *Chapman rearrangement of* 2, 4-di-(4-carbethoxy-2, 6-dimethoxyphenoxy)-6-methylpyrimidine **(3e)** under microwave irradiation (900W) in 18 minutes.

6. Synthesis of 1, 3-di-(2-ethoxyphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4f)** as described in **Example-6** by *Chapman rearrangement* of 2, 4-di-(2-ethoxyphenoxy)-6-methylpyrimidine **(3f)** under microwave irradiation (900W) in 20 minutes.

7. Synthesis of 1, 3-di-(4-chloro-3, 5-dimethylphenyl)-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4g)** as described in **Example-7** by *Chapman rearrangement* of 2, 4-di-(4-chloro-3, 5-dimethylphenoxy)-6-methylpyrimidine **(3g)** under microwave irradiation (900W) in 15 minutes.

8. Synthesis of 1, 3-diphenyl-(1H, 3H)-6-methylpyrimidine-2, 4-dione **(4h)** as described in **Example-8** by *Chapman rearrangement* of 2, 4-diphenoxy-6-methylpyrimidine **(3h)** under microwave irradiation (900W) in 16 minutes.

9. 2, 4-diaryloxy-6-methylpyrimidines **(3a-3h)** underwent facile *Chapman rearrangement* under microwave irradiation to afford 1, 3- disubstituted-6-methylpyrimidine-2, 4-diones **(4a-4j)** which provides a simpler and environmental friendly procedure. Thus, microwave is a convenient way towards the goal of green, sustainable chemistry, and is strongly recommended to use in organic preparations.

6. DATE: 29th **October 2013**

SIGNATURES:

Dr. M. M. V. Ramana **Dr. Sanjay C. Pawar**