7. ABSTRACT OF THE INVENTION:

This invention describes the synthesis of 1, 3-diaryl-(1H, 3H)-pyrimidine-2, 4-diones by subjecting corresponding 2, 4-diaryloxypyrimidines to *Chapman rearrangement* under microwave irradiation.

5. CLAIMS:

We Claim:

1. Synthesis of 1, 3-di-(4-carbmethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4a)** as described in **Example-1** by *Chapman rearrangement* of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** under microwave irradiation (900W) in 15 minutes.
2. Synthesis of 1, 3-di-(2-carbethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4b)** as described in **Example-2** by *Chapman rearrangement* of 2, 4-di-(2-carbethoxyphenoxy)-pyrimidine **(3b)** under microwave irradiation (900W) in 13 minutes.
3. Synthesis of 1, 3-diphenyl-(1H, 3H)-pyrimidine-2, 4-dione **(4c)** as described in **Example-3** by *Chapman rearrangement* of 2, 4-diphenoxypyrimidine **(3c)** under microwave irradiation (900W) in 12 minutes.
4. Synthesis of 1, 3-di-(1-naphthyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4d)** as described in **Example-4** by *Chapman rearrangement* of 2, 4-di-(1-naphthoxy)-pyrimidine **(3d)** under microwave irradiation (900W) in 13 minutes.
5. Synthesis of 1, 3-di-(4-carbethoxy-2, 6-dimethoxy phenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4e)** as described in **Example-5** by *Chapman rearrangement* of 2, 4-di-(4-carbethoxy-2, 6-dimethoxyphenoxy)-pyrimidine **(3e)** under microwave irradiation (900W) in 18 minutes.
6. Synthesis of 1, 3-di-(2-ethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4f)** as described in **Example-6** by *Chapman rearrangement* of 2, 4-di-(2-ethoxyphenoxy)-pyrimidine **(3f)** under microwave irradiation (900W) in 15 minutes.
7. Synthesis of 1, 3-di-(4-ethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4g)** as described in **Example-7** by *Chapman rearrangement* of 2, 4-di-(4-ethoxyphenoxy)-pyrimidine **(3g)** under microwave irradiation (900W) in 13 minutes.
8. Synthesis of 1, 3-di-(4-chloro-3, 5-dimethylphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4h)** as described in **Example-8** by *Chapman rearrangement* of 2, 4-di-(4-chloro-3, 5-dimethylphenoxy)-pyrimidine **(3h)** under microwave irradiation (900W) in 15 minutes.
9. Synthesis of 1, 3-di-(2-methylphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4i)** as described in **Example-9** by *Chapman rearrangement* of 2, 4-di-(2-methylphenoxy)-pyrimidine **(3i)** under microwave irradiation (900W) in 15 minutes.
10. 2, 4-diaryloxy pyrimidines **(3a-3i)** underwent facile *Chapman rearrangement* to afford 1, 3-diarylpyrimidine-2, 4-diones **(4a-4i)** under microwave irradiation which provides a simpler and environmental-friendly procedure under green chemistry conditions having promising future in industries.

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

SIGNATURE:-

Mramana

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

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4. DESCRIPTION:

Title:

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

Field of invention:

This invention describes the synthesis of 1, 3-diaryl-(1H, 3H)-pyrimidine-2, 4-diones by subjecting corresponding 2, 4-diaryloxypyrimidines to *Chapman rearrangement* under microwave irradiation.

Background of invention and Prior art:

Pyrimidinediones have attracted immense interest in organic synthesis as well as as pharmaceutical industry, as they exhibit attractive pharmacological profiles. (D. Cole, A. J. Foster, S. W. Freeman, P. E. Murray, I. J. Stanford, *Anti-Cancer Drug Das,* **1999,** 14,383; S. Senda, K. J. Hirota, *Med Chem,* **1972**, 15, 471) Additionally, they act as important intermediates for the synthesis of pyrimidine bases that also possess similar therapeutic profiles. A number of N-substituted pyrimidinediones possessing good biological activities are well known in prior arts including US 6232318 B1, EP 1924567, US 6759415, US 6420385, US 6410729. Uramustine, 5-[N, N'-bis (2'-chloroethyl) amino]-pyrimidine-2, 4-dione are used orally in the treatment of several leukemias, and 5-nitro-(1H, 3H)-pyrimidine-2, 4-dione derivatives exhibit macrophage growth inhibition. (A. Copik, J. Suwinski, K. Walczak, J. Bronkowska, Z. Czuba and W. Krol, *Nucleosides, Nucleotides, Nucleic acids, Res.*, **2002**, 21, 377-383) 1-Aryl-5-substituted pyrimidine-2, 4-diones 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; A. Gondela and K. Walczak, *Tetrahedron Asymmetry,* **2005**, 16, 2107-2112) Pyrimidinedione derivatives are used for treating or preventing metabolic disorders, neurological disorders, hematological diseases, cancer, inflammation, respiratory diseases, gastroenterological diseases, diabetic complications, an obesity-related disorders and non-alcoholic fatty liver diseases (WO2008127591 A2). Some pyrimidinedione derivatives have got agricultural importance since they are well known for their use as pesticides, antiphotosynthetic herbicides, and insecticides. Such materials are well known in the prior arts including WO 1998008824 A1, WO 0002866, US 5344812, US 5399543, US 5674810, US 5726124, US 5300477, WO 99/21837, JP 03287585 A2, WO 9952906.

1-Arylpyrimidine-2, 4-diones have been synthesized from substituted ureidopropanoic acids or 1-acryloyl-3-arylureas. (N. W. Gabel and S. B. Binkley, *J. Org. Chem.*, **1958**, 23, 643-645). 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; S. A. Jacobsen, S.Rodbotten and T. Benneche, *J. Chem. Soc., Perkin Trans.* 1, **1999**, 3262-3268).

Method of preparation of most of the above reported 1, 3-diaryl-(1H, 3H)-pyrimidine-2,4-diones suffers from one of the disadvantages such as extended time, harsh reaction conditions, tedious preparation procedure, use of solvents etc. Thus, there is scope for the synthesis of them by simple and eco-friendly method. The present invention describes their synthesis in two steps through *Chapman rearrangement* under microwave irradiation in absence of solvent in second step.

The use of microwave irradiation in organic synthesis has become increasingly popular within the pharmaceutical and academic arenas. (Loupy, A., *Microwaves in Organic Synthesis*, Wiley-VCH: Weinheim, **2002**.) Presently, thermally driven organic transformations take place by either of two ways: 1. Conventional heating 2. Microwave accelerated heating. The first is a slow and inefficient method for transferring energy into the reacting system since the reactants are slowly activated where heat energy passes first through the walls of the vessel in order to reach the solvent and reactants. In the second way, microwaves couple directly with the molecules of the entire reaction mixture, leading to a rapid rise in temperature. This dramatically decreases reaction time, clean work up and better reaction yield. (Hayes, B. L. *Microwave Synthesis: Chemistry at theSpeed of Light*; CEM Publishing: Matthews, NC, **2002**.)

Description of the invention:

A series of 1, 3-diaryl-(1H, 3H)-pyrimidine-2, 4-dione derivatives have been synthesized by subjecting 2, 4-diaryloxy pyrimidines to *Chapman rearrangement* under microwave irradiation.

The main objective of the present invention is to synthesize compounds having few steps of reactions, less reaction time, minimum use of solvents, clean work up, and better reaction yield. The present invention is successful in attaining all the above objectives providing a simpler and environmental friendly alternative for the conventional procedures.

The thermal conversion of aryl N-arylbenzimidates to N-aroyldiphenylamines is known as the *Chapman rearrangement*. (Chapman, *J. Chem. Soc.* **1925**, 569; Chapman, *J. Chem. Soc.* **1927**, 1743)

For this purpose, 1H, 3H-pyrimidine-2, 4-dione was utilized as the starting substrate. This on chlorination followed by condensation with various phenols yielded the respective aryloxy derivatives. These were subjected to *Chapman rearrangement* under microwave irradiation to afford the corresponding 1, 3-diaryl-(1H, 3H)-pyrimidine-2, 4-diones. **(Scheme)**

Examples:

2, 4-dichloropyrimidine **(1)** has been synthesized as per literature procedure from 1H, 3H-pyrimidine-2, 4-dione. (Whittaker, N., *J. Am. Chem. Soc.*, **1965**, *87*, 4569)

Example-1: Preparation of 1, 3-di-(4-carbmethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione (4a).

Step A- Preparation of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine (3a).

A mixture of 2, 4-dichloropyrimidine (1) (0.03M), 4-carbmethoxyphenol **(2a)** (0.06M) and K_2CO_3 (0.06M) in dry acetone (50 ml) was slowly refluxed for 4.5 hours under dry conditions. After completion of the reaction (TLC), acetone was recovered by flash distillation. The reaction mass was cooled to room temperature and quenched in water (50ml) under stirring. The heterogeneous solution was extracted in dichloromethane (2 x 25ml) followed by washing with 5% NaOH solution (2 x 25ml). The combined extracts were given water washing (2 x 25ml) and dried over sodium sulphate. Recovery of solvent followed by purification afforded viscous oil (Yield: 62%) of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)**.

IR (KBr, cm^{-1}): 1215, 1253, 1350, 1606, 1720, 2978-3032.

^{1}H NMR (300 MHz, $CDCl_3$): δ 3.9(s, 6H), 6.4-7.6(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 52-53.5, 125-172, 168.5-172.

Molecular formula: $C_{20}H_{16}N_2O_6$. Elemental analysis: Calculated: C (63.16%), H (4.21%), N (7.37%). Found: C (63.21%), H (4.28%), N (7.42%).

HRMS: *m/z* cal. mass for $C_{20}H_{16}N_2O_6$ $[M+H]^+$ =380.3528, obs. mass $[M+H]^+$ = 380.3562.

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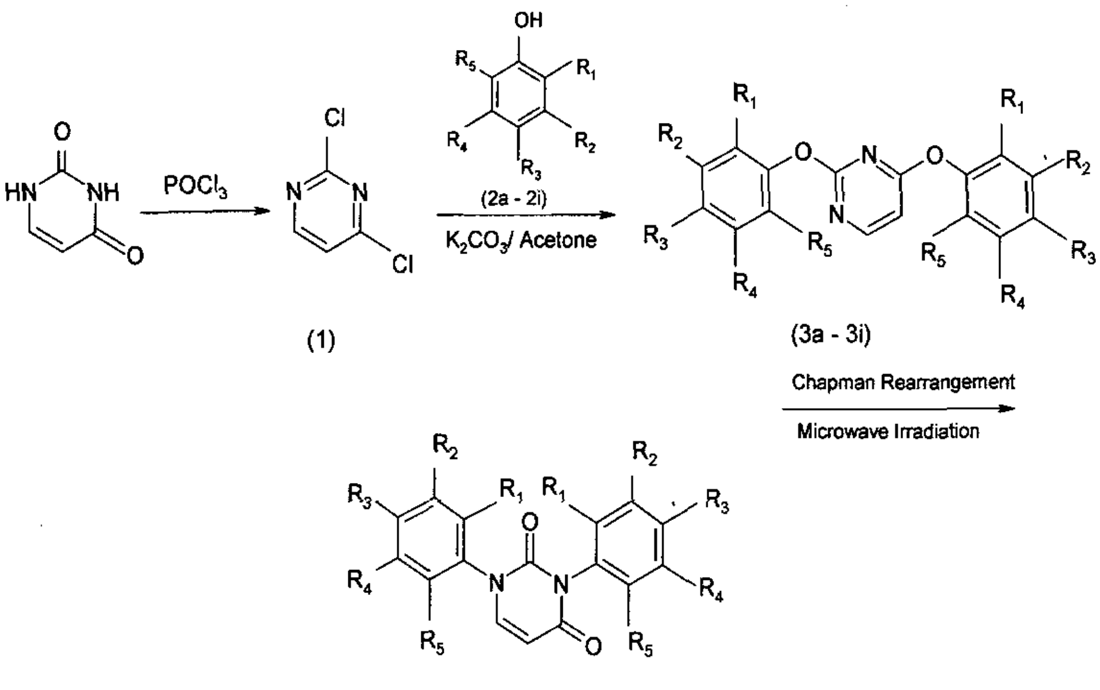
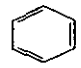
Scheme:



(4a - 4i)

Examples	Compounds	R_1	R_2	R_3	R_4	R_5
1.	2a, 3a, 4a	H	H	$COOCH_3$	H	H
2.	2b, 3b, 4b	$COOC_2H_5$	H	H	H	H
3.	2c, 3c, 4c	H	H	H	H	H
4.	2d, 3d, 4d	H	H	H		
5.	2e, 3e, 4e	OCH_3	H	$COOC_2H_5$	H	OCH_3
6.	2f, 3f, 4f	OC_2H_5	H	H	H	H
7.	2g, 3g, 4g	H	H	OC_2H_5	H	H
8.	2h, 3h, 4h	H	CH_3	Cl	CH_3	H
9.	2i, 3i, 4i	H	CH_3	H	H	H

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Step B- Preparation of compound 4a.

In a flask, equipped with water condenser 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** (0.02M) was irradiated (900W) in microwave reactor for 15 minutes. The reaction mass was cooled to room temperature and treated with ligroin (50 ml). The solid obtained was filtered and recrystallised from ligroin to give white crystals of compound **4a** (Yield: 76%). (m.p.: 130°C)

IR (KBr, cm^{-1}): 1210, 1358, 1606, 1635, 1688, 1700, 1715, 2990-3278.

^1H NMR (300 MHz, $CDCl_3$): δ 3.5(s, 6H), 6-7.2(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 52.5-53.5, 127.5-158, 98, 141, 152.5, 164, 165-167.5.

Molecular formula: $C_{20}H_{16}N_2O_6$. Elemental analysis: C (63.16%), H (4.21%), N (7.37%). Found: C (63.07%), H (4.25%), N (7.41 %).

HRMS: *m/z* cal. mass for $C_{20}H_{16}N_2O_6$ $[M+H]^+$ =380.3528, obs. mass $[M+H]^+$ = 380.3503.

Example-2: Preparation of 1, 3-di-(2-carbethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione (4b).

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

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloropyrimidine **(1)** and 2-carbethoxyphenol **(2b)** instead of 4-carbmethoxyphenol **(2a)** whereby **3b** was obtained as viscous oil (Yield: 64%) after purification.

IR (KBr, cm^{-1}): 1242, 1358, 1609, 1708, 2978-2985.

^1H NMR (300 MHz, $CDCl_3$): δ 1.4(t, J = 7.3 Hz, 6H), 4.1(q, J = 7.6Hz, 4H), 6.3-7.0(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 14.5-15, 57.5-63, 121-168, 169-172.

Molecular formula: $C_{22}H_{20}N_2O_6$. Elemental analysis: Calculated: C (64.71%), H (4.90%), N (6.86%). Found: C (64.78%), H (4.93%), N (6.93%).

HRMS: *m/z* cal. mass for $C_{22}H_{20}N_2O_6$ $[M+H]^+$ =408.4061, obs. mass $[M+H]^+$ = 408.4078.

Step B- Preparation of compound 4b.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(2-carbethoxyphenoxy)-pyrimidine **(3b)** instead of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** was irradiated (900W) in microwave reactor for 13 minutes, whereby **4b** was obtained as white crystals (Yield: 69%) after purification. (m.p.:123°C)

IR (KBr, cm^{-1}): 1200, 1341, 1600, 1640, 1680, 1691, 1700, 2854-2955.

^1H NMR (300 MHz, $CDCl_3$): δ 1.3(t, J=6.9 Hz, 6H), 4.6(q, J=7.4Hz, 4H), 6.2-7.4(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 13- 14.5, 62-64, 108.5, 126-162.5, 141, 153.5, 168, 169.5-175.

Molecular formula: $C_{22}H_{20}N_2O_6$. Elemental analysis: Calculated: C (64.71%), H (4.90%), N (6.86%). Found: C (64.76%), H (4.87%), N (6.90%).

HRMS: *m/z* cal. mass for $C_{22}H_{20}N_2O_6$ $[M+H]^+$=408.4061, obs. mass $[M+H]^+$ = 408.4013.

Example-3: Preparation of 1, 3-diphenyl-(1H, 3H)-pyrimidine-2, 4-dione (4c).

Step A- Preparation of 2, 4-diphenoxypyrimidine (3c).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloropyrimidine **(1)** and phenol **(2c)** instead of 4-carbmethoxyphenol **(2a)**, whereby **3c was** obtained as crystals (Yield: 74%) after purification. (m.p.: 111°C) (Ghosh, S. K.; Dolly, R. S.; Mukherjee, M. K. *J. Med. Chem.* **1968**, *11 (6)*, 1237-8)

Step B- Preparation of compound 4c.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-diphenoxypyrimidine **(3c)** instead of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** was irradiated (900W) in microwave reactor for 12 minutes, whereby **4c** was obtained as white crystals (Yield: 82%) after purification. (m.p.:142°C) (Hall, S.C.; *J. Chem. Soc.* **1942**, 64)

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

Step A- Preparation of 2, 4-di-(1-naphthoxy)-pyrimidine (3d).

It was prepared as described in **Step A** of Example-1 by using 2, 4-dichloropyrimidine **(1)** and 1-naphthol **(2d)** instead of 4-carbmethoxyphenol **(2a)**, whereby **3d** was obtained as cream colored solid (Yield: 62%) after purification. (m.p.:129°C) (Proffit, E.; Raddatz, H. *Arch. Pharm.* 1962, *295*, 649-62).

Step B- Preparation of compound 4d.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(1-naphthoxy)-pyrimidine **(3d)** instead of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** was irradiated (900W) in microwave reactor for 13 minutes, whereby **4d** was obtained as an oil (Yield: 71%) after purification.

IR (KBr, cm^{-1}): 1327, 1601, 1649, 1685, 1691.

^{1}H NMR (300 MHz, $CDCl_3$): δ 6.2-7.3(m, 16H).

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

Molecular formula: $C_{24}H_{16}N_2O_2$. Elemental analysis: C (79.12%), H 4.40%), N (7.69%). Found: C (79.19%), H (4.49%), N (7.76 %).

HRMS: *m/z* cal. mass for $C_{24}H_{16}N_2O_2$ $[M+H]^+$ = 364.3982, obs. mass $[M+H]^+$ =364.3959.

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

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

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloropyrimidine **(1)** and ethyl-4-hydroxy-3, 5-dimethoxybenzoate (ethyl syringate) **(2e)** instead of 4-carbmethoxyphenol **(2a),** whereby **3e** was obtained as viscous oil (Yield: 59%) after purification.

IR (KBr, cm^{-1}): 1215, 1253, 1350, 1606, 1720, 2978-3032.

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.5(t, J=7.1 Hz, 6H), 3.7(s, 12H), 4.4(q, J=7.6 Hz, 4H), 6.1-7.3 (m, 6H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 20.5-21.5, 54, 56, 56.5-63, 165-170.5, 126-172.5.

Molecular formula: $C_{26}H_{28}N_2O_{10}$. Elemental analysis: Calculated: C (59.09%), H (5.30%), N (5.30%). Found: C (59.14%), H (5.36%), N (5.19 %).

HRMS: *m/z* cal. mass for $C_{26}H_{28}N_2O_{10}$ $[M+H]^+$=528.5105, obs. mass $[M+H]^+$ = 528.5131

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)-pyrimidine **(3e)** instead of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine (3a) was irradiated (900W) in microwave reactor for 18 minutes, whereby **4e** was obtained as an oil (Yield: 71%) after purification.

IR (KBr, cm^{-1}): 1205, 1249, 1350, 1606, 1642, 1681, 1695, 1710, 2990-3000.

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.7(t, J=7.3 Hz, 6H), 3.9(s, 12H), 4.4(q, J=7.9 Hz, 4H), 6.0-7.2 (m, 6H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 21-22.5, 53-54.5, 60-62.5, 127-157.5, 101.5, 142, 152.5, 161, 162.5-166.5.

Molecular formula: $C_{26}H_{28}N_2O_{10}$. Elemental analysis: Calculated: C (59.09%), H (5.30%), N (5.30%). Found: C (59.01%), H (5.35%), N (5.38%).

HRMS: *m/z* cal. mass for $C_{26}H_{28}N_2O_{10}$ $[M+H]^+$=528.5105, obs. mass $[M+H]^+$ = 528.5170

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

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

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloropyrimidine **(1)** and 2-ethoxyphenol **(2f)** instead of 4-carbmethoxyphenol **(2a)**, whereby **3f was** obtained as reddish crystals (Yield: 63%) after purification. (m.p.: 102°C)

IR (KBr, cm^{-1}): 1251, 1355, 1605, 2854-3120.

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.2(t, J=6.7 Hz, 6H), 3.9(q, J=7.4 Hz, 4H), 6.2-7.2(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 13.5-15, 57.5-64, 125-168.5.

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Molecular formula: $C_{20}H_{20}N_2O_4$. Elemental analysis: Calculated: C (68.18%), H (5.68%), N (7.95%). Found: C (68.24%), H (5.73%), N (8.03%).

HRMS: *m/z* cal. mass for $C_{20}H_{20}N_2O_4$ $[M+H]^+$=352.3857, obs. mass $[M+H]^+$= 352.3808.

Step B- Preparation of compound 4f.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(2-ethoxyphenoxy)-pyrimidine **(3f)** instead of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** at was irradiated (900W) in microwave reactor for 15 minutes, whereby **4f** was obtained as an oil (Yield: 72%) after purification.

IR (KBr, cm^{-1}): 1241, 1358, 1618, 1644, 1688, 1695, 2997-3038.

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.9(t, J=6.9 Hz , 6H), 4.1(q, J=7.8Hz, 4H), 6.1-6.9(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 14-15.5, 54-56.5, 101, 127.5-160, 141.5, 154.5, 169.

Molecular formula: $C_{20}H_{20}N_2O_4$. Elemental analysis: Calculated: C (68.18%), H (5.68%), N (7.95%). Found: C (68.07%), H 5.75%), N (7.90%).

HRMS: *m/z* cal. mass for $C_{20}H_{20}N_2O_4$ $[M+H]^+$=352.3857, obs. mass $[M+H]^+$= 352.3832.

Example-7: Preparation of 1, 3-di-(4-ethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione (4g).

Step A- Preparation of 2, 4-di-(4-ethoxyphenoxy)-pyrimidine (3g).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloropyrimidine **(1)** and 4-ethoxyphenol **(2g)** instead of 4-carbmethoxyphenol **(2a),** whereby **3g** was obtained as colourless crystals (Yield: 60%) after purification. (m.p.: 95°C)

IR (KBr, cm^{-1}): *1230, 1351, 1612, 2992-3025.*

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.3(t, J=7.1 Hz, 6H), 4.4(q, J=7.9 Hz, 4H), 6.1-7.2(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 17.5-18.5, 58-63, 124-173.

Molecular formula: $C_{20}H_{20}N_2O_4$. Elemental analysis: Calculated: C (68.18%), H (5.68%), N (7.95%). Found: C (68.08%), H (5.56%), N (7.83%).

HRMS: *m/z* cal. mass for $C_{20}H_{20}N_2O_4$ $[M+H]^+$=352.3857, obs. mass $[M+H]^+$= 352.3881.

Step B- Preparation of compound 4g.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(4-ethoxyphenoxy)-pyrimidine **(3g)** instead of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** was irradiated (900W) in microwave reactor for 13 minutes, whereby **4g** was obtained as an oil (Yield: 67%) after purification.

IR (KBr, cm^{-1}): 1238, 1340, 1611, 1633, 1691, 1692, 2923-3010.

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.5(t, J=6.8 Hz, 6H); 4.5(q, J=7.9Hz, 4H), 6-7.3(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 16.5-17.5, 58.5-61, 102.5, 128-149, 141, 153, 165.5.

Molecular formula: $C_{20}H_{20}N_2O_4$. Elemental analysis: Calculated: C (68.18%), H (5.68%), N (7.95%). Found: C (68.24%), H 5.73%), N (8.02%).

HRMS: *m/z* cal. mass for $C_{20}H_{20}N_2O_4$ $[M+H]^+$ =352.3857, obs. mass $[M+H]^+$ = 352.3823.

Example-8: Preparation of 1, 3-di-(4-chloro-3, 5-dimethylphenyl)-(1H,3H)-pyrimidine-2, 4-dione (4h).

Step A- Preparation of 2, 4-di-(4-chloro-3, 5-dimethylphenoxy)-pyrimidine (3h).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloropyrimidine **(1)** and 4-chloro-3,5-dimethylphenol **(2h)** instead of 4-carbmethoxyphenol **(2a),** whereby **3h** was obtained as colorless crystals (Yield: 69%) after purification. (m.p.: 107°C)

IR (KBr, cm^{-1}): 780, 1251, 1356, 1608, 2906-2987.

^1H NMR (300 MHz, $CDCl_3$): δ 1.4(s, 12H), 6.2-7.5(m, 6H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 18-21.5, 125-172.5.

Molecular formula: $C_{20}H_{18}N_2O_2Cl_2$. Elemental analysis: Calculated: C (61.70%), H (4.63%), N (7.20%), Cl (18.25%). Found: C (61.76%), H (4.57%), N (7.27%), Cl (18.11%).

HRMS: *m/z* cal. mass for $C_{20}H_{18}N_2O_2Cl_2$ $[M+H]^+$ =389.2773, obs. mass $[M+H]^+$ = 389.2708.

Step B- Preparation of compound 4h.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(4-chloro-3, 5-dimethylphenoxy)-pyrimidine **(3h)** instead of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** was irradiated (900W) in microwave reactor for 15 minutes, whereby **4h** was obtained as an oil (Yield: 72%) after purification.

IR (KBr, cm^{-1}): 788, 1349, 1612, 1621, 1689, 1708, 2976-3046.

^1H NMR (300 MHz, $CDCl_3$): δ 1.4(s, 12H), 6.1-7.3(m, 6H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 16.5-19, 102, 127-135, 146, 155, 165.5.

Molecular formula: $C_{20}H_{18}N_2O_2Cl_2$. Elemental analysis: Calculated: C (61.70%), H (4.63%), N (7.20%), Cl (18.25%). Found: C (61.74%), H (4.67%), N (7.24%), Cl (18.29%).

HRMS: *m/z* cal. mass for $C_{20}H_{18}N_2O_2Cl_2$ $[M+H]^+$ =389.2773, obs. mass $[M+H]^+$ = 389.2774.

Example-9: Preparation of 1, 3-di-(2-methylphenyl)-(1H, 3H)-pyrimidine-2, 4-dione (4i).

Step A- Preparation of 2, 4-di-(2-methylphenoxy)-pyrimidine (3i).

It was prepared as described in **Step A** of **Example-1** by using 2, 4-dichloropyrimidine **(1)** and 2-methyl phenol **(2i)** instead of 4-carbmethoxyphenol **(2a),** whereby **3i was** obtained as white crystals (Yield: 71%) after purification. (m.p.: 93°C) (Sotiropoulos, Jean, Lamazonere, Anne M. *CR. Acad. Sci. Ser.C;*. **1970**, *271 (25)*, 1592-5)

Step B- Preparation of compound 4i.

It was prepared as described in **Step B** of **Example-1** by using 2, 4-di-(2-methylphenoxy)-pyrimidine **(3i)** instead of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** was irradiated (900W) in microwave reactor for 15 minutes, whereby **4i** was obtained as an oil (Yield: 64%) after purification.

IR (KBr, cm^{-1}): 1356, 1619, 1631, 1683, 1697, 2923-3034.

^{1}H NMR (300 MHz, $CDCl_3$): δ 1.6(s, 6H), 6.2-7.6(m, 10H).

^{13}C NMR (75 MHz, $CDCl_3$): δ 17-18.5, 102, 123.5, 126-138.5, 154, 164.5.

Molecular formula: $C_{18}H_{16}N_2O_2$. Elemental analysis: Calculated: C (73.97%), H (5.48%), N (9.59%). Found: C (73.92%), H (5.39%), N (9.64%).

HRMS: *m/z* cal. mass for $C_{18}H_{16}N_2O_2$ $[M+H]^+$ =292.3335, obs. mass $[M+H]^+$ = 292.3364.

5. CLAIMS:

We Claim:

1. Synthesis of 1, 3-di-(4-carbmethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4a)** as described in **Example-1** by *Chapman rearrangement* of 2, 4-di-(4-carbmethoxyphenoxy)-pyrimidine **(3a)** under microwave irradiation (900W) in 15 minutes.
2. Synthesis of 1, 3-di-(2-carbethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4b)** as described in **Example-2** by *Chapman rearrangement* of 2, 4-di-(2-carbethoxyphenoxy)-pyrimidine **(3b)** under microwave irradiation (900W) in 13 minutes.
3. Synthesis of 1, 3-diphenyl-(1H, 3H)-pyrimidine-2, 4-dione **(4c)** as described in **Example-3** by *Chapman rearrangement* of 2, 4-diphenoxypyrimidine **(3c)** under microwave irradiation (900W) in 12 minutes.
4. Synthesis of 1, 3-di-(1-naphthyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4d)** as described in **Example-4** by *Chapman rearrangement* of 2, 4-di-(1-naphthoxy)-pyrimidine **(3d)** under microwave irradiation (900W) in 13 minutes.
5. Synthesis of 1, 3-di-(4-carbethoxy-2, 6-dimethoxy phenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4e)** as described in **Example-5** by *Chapman rearrangement* of 2, 4-di-(4-carbethoxy-2, 6-dimethoxyphenoxy)-pyrimidine **(3e)** under microwave irradiation (900W) in 18 minutes.
6. Synthesis of 1, 3-di-(2-ethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4f)** as described in **Example-6** by *Chapman rearrangement* of 2, 4-di-(2-ethoxyphenoxy)-pyrimidine **(3f)** under microwave irradiation (900W) in 15 minutes.
7. Synthesis of 1, 3-di-(4-ethoxyphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4g)** as described in **Example-7** by *Chapman rearrangement* of 2, 4-di-(4-ethoxyphenoxy)-pyrimidine **(3g)** under microwave irradiation (900W) in 13 minutes.
8. Synthesis of 1, 3-di-(4-chloro-3, 5-dimethylphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4h)** as described in **Example-8** by *Chapman rearrangement* of 2, 4-di-(4-chloro-3, 5-dimethylphenoxy)-pyrimidine **(3h)** under microwave irradiation (900W) in 15 minutes.
9. Synthesis of 1, 3-di-(2-methylphenyl)-(1H, 3H)-pyrimidine-2, 4-dione **(4i)** as described in **Example-9** by *Chapman rearrangement* of 2, 4-di-(2-methylphenoxy)-pyrimidine **(3i)** under microwave irradiation (900W) in 15 minutes.
10. 2, 4-diaryloxy pyrimidines **(3a-3i)** underwent facile *Chapman rearrangement* to afford 1, 3-diarylpyrimidine-2, 4-diones **(4a-4i)** under microwave irradiation which provides a simpler and environmental-friendly procedure under green chemistry conditions having promising future in industries.

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

SIGNATURE:-

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

29 OCT 2013